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# **CADMIUM IN THE EUROPEAN COMMUNITY**

**Technical Report**

Prepared by:

**MONITORING AND ASSESSMENT RESEARCH CENTRE**  
Chelsea College, University of London

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# **Cadmium in the European Community: a prospective assessment of sources, human exposure and environmental impact**

by Malcolm Hutton

**A Technical Report (1982)**

Prepared by:  
The Monitoring and Assessment Research Centre  
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## SUMMARY

This study quantifies the sources of cadmium in the European Community (EC), reviews the concentrations found in the physical environment and its biotic components, and assesses current and future human exposure in the region. No attempt has been made to evaluate cadmium toxicity data generated from human studies or experimentally dosed animals. The risk of adverse health effects in humans is considered only in relation to the magnitude of exposure.

A large part of the report is concerned with quantifying the sources of cadmium in the EC and assessing the relative significance of such inputs to the environmental compartments, air, land and water. These findings have been critically compared with previous estimates of cadmium inputs in the region.

The steel industry and waste incineration, followed by volcanic action and zinc production, are estimated to account for the largest emissions of atmospheric cadmium in the community. Waste disposal results in the single largest input of cadmium to land; the quantity of cadmium associated with this source is greater than the total from the four other major sources, coal combustion, iron and steel production, phosphate fertilizer manufacture and use, and zinc production. It has not been possible to characterize fully cadmium inputs to aquatic systems, but of the sources considered, the manufacture of cadmium-containing articles accounts for the largest aquatic discharge, followed by phosphate fertilizer manufacture and zinc production.

Predictions of future cadmium discharges in the EC indicate that by the year 2000 there will be significant increases in atmospheric inputs from zinc production, iron and steel production and sewage sludge incineration. It is also forecast that iron and steel production together with coal combustion will be the major sources responsible for large increases in cadmium discharged to land by the year 2000.

Published information on cadmium concentrations in those media associated with human exposure is surveyed and representative values selected for background and contaminated localities. Attention is also paid to the accumulation and significance of cadmium in plants,

invertebrates and vertebrates and the mobility of cadmium in terrestrial food chains.

The sources of cadmium exposure in the general population of the EC are examined and the relative contribution from different routes evaluated. Attention is paid to the problems associated with the estimation of dietary intake and expressing exposure and uptake on an average basis.

A modelling technique is developed to examine the consequences of cadmium inputs to agricultural land upon future dietary intake of cadmium in the EC. This technique is also employed to assess the possible impact of two hypothetical control strategies upon future cadmium exposure. It is forecast that there will be significant increases in dietary intake of cadmium in the future. By the year 2000, it is estimated that average intake values will increase by at least 50 per cent of present-day values.

The removal of cadmium inputs associated with phosphate fertilizer use is predicted to result in dietary intakes 100 years from now which are only slightly greater than present-day values. In contrast, the cessation of cadmium emissions from iron and steel production is estimated to have minimal influence on the predicted increases in future dietary intake.

## **FOREWORD**

This document is the modified version of a report submitted to the Commission of the European Communities under Contract Number 333-ENV U.K. The work on which this report is based was commissioned by the Environment, Raw Materials and Material Technologies Research Programmes, Directorate General for Science, Research and Development. Greece became a Member of the European Community after this study was begun and is therefore not considered in this report. The views expressed in the report do not necessarily reflect those of the Commission.

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## **1 Sources of cadmium in the EC**

### **1.1 Introduction**

This section is concerned with determining the magnitude of present-day cadmium discharges from various sources in the EC. The findings are compared with two previous inventories for the region (Rauhut 1978; Van Wambeke 1979). Additionally, this study also provides predictions of cadmium discharges in the EC for 1990 and 2000.

The data provided in this section were obtained from numerous sources and in many instances requests for information from industry were made when published studies were unavailable. In some cases, information on cadmium emissions from a particular source had to be derived from studies carried out in countries outside the EC.

Cadmium inputs were considered for the three environmental compartments, air, land and water. There is a paucity of information regarding the magnitude of cadmium discharges to water bodies. For this reason, the inventory of cadmium discharges to water is incomplete.

### **1.2 Natural sources**

The quantities of cadmium derived from natural sources are difficult to estimate, largely because there have been very few measurements of the emission or mobilization rates from such processes. Sea sprays, wind-blown dusts and forest fires all release cadmium to the atmosphere but even on a global scale the amounts involved are considered to be relatively small (Nriagu 1979). Moreover, the most important of the above processes, wind-blown dust erosion, occurs mainly in desert areas and these are absent from the study region.

Recently, attention has been paid to the importance of plants as emitters of trace metals to the atmosphere (Beauford, Barber and Barringer 1977). This process may well be a significant source of atmospheric cadmium, but to date insufficient data exist to allow an estimate of the quantity involved.

Volcanic action is considered to be the major natural source of cadmium in the atmosphere. This is related to the very large quantities of particulate matter emitted by volcanoes, together with high enrichment of cadmium in volcanic aerosols. The enrichment derives from the condensation of cadmium previously vaporized by the elevated temperatures associated with volcanic activity.

One of the few investigations into cadmium emissions from volcanoes examined Mount Etna in Sicily (Buat-Ménard and Arnold 1978). This study estimated that  $2.8 \times 10^{-2} \text{ t}^* \text{ d}^{-1}$  or about  $10 \text{ t y}^{-1}$  cadmium was discharged into the atmosphere from this volcano. It should be noted that the estimate is probably lower than the total discharge of cadmium as only particulate matter was sampled and the quantity of cadmium in the gaseous state was not measured. Indeed, more recent investigations by Buat-Ménard (pers. comm.) indicate that the annual cadmium emission from Mount Etna lies within the range of 10–40 t. No published information exists on cadmium emissions from other active volcanoes in Europe but these are far less important than Mount Etna in terms of the quantity of particulate matter released.

The weathering of crustal materials releases cadmium to soils and aquatic systems. This process plays a significant rôle in the global cadmium cycle but only rarely results in elevated concentrations in any environmental compartment. Black shales from the Carboniferous era are an exception as they contain up to  $100 \text{ mg kg}^{-1}$  cadmium and soils derived from these deposits also contain elevated levels (Holmes 1975). Similarly, non-ferrous ore bodies which contain cadmium will contaminate adjacent water courses and soils. However, the mining of such deposits results in a much greater mobilization of the metal.

### 1.3 The production of non-ferrous metals

Cadmium is a constituent of the ores of zinc, lead and copper. Consequently, primary production of these metals invariably results in discharges of cadmium to the environment. Cadmium emissions also occur from secondary non-ferrous metal production; these result from impurities in the scrap metal charge.

#### 1.3.1 Zinc and cadmium

There are two distinct processes which are used in the primary production of zinc and cadmium—thermal smelting or electrolytic extraction. Both processes share a common initial step, the roasting of ore concentrates to convert zinc sulphide to zinc oxide. The next stage in the thermal process, sintering, employs much higher temperatures which result in the vaporization of the major part of the cadmium present in the ore.

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\* Metric tons or tonnes ( $10^3 \text{ kg}$ ) are used throughout this report.

The sinter is subsequently smelted and zinc and cadmium oxide are reduced by carbon monoxide, produced by the addition of coke. Both cadmium and zinc are vaporized and collected in condensers. Further purification of the zinc is carried out by distillation. The cadmium present in the dusts originating from the roasting and sintering stages is recovered and purified in a separate operation.

In the electrolytic process, the roasted desulphurized ore is leached with sulphuric acid to produce zinc sulphate solution. Cadmium and other impurities are precipitated by the addition of zinc dust. The cadmium-rich precipitate and the zinc sulphate solution are then processed separately in electrolytic cells.

### *Emission sources*

Emission sources from zinc production have been characterized previously (Rauhut 1978). Thermal facilities emit much larger quantities of cadmium to the atmosphere, as a consequence of the higher temperatures employed in the roasting, sintering, and smelting operations. Nevertheless, the use of modern control technology at all stages results in the reduction of airborne cadmium emissions, especially from the roasting and smelting operations.

Both electrolytic and thermal processes discharge cadmium-rich dusts into the atmosphere during the delivery and handling of the zinc ores. This emission source consists of relatively large particles and, in contrast to stack emissions, these will remain airborne for only a short time and will be deposited close to the plant.

Liquid wastes containing cadmium arise primarily from the use of wet gas purification devices. In electrolytic works, this water can be recycled to the production process. Waste waters from thermal plants pose a more difficult problem and they are often discharged to adjacent water systems after undergoing a cleaning process.

The major solid waste requiring disposal from thermal facilities is smelter slag. This contains low cadmium concentrations and does not appear to pose a disposal problem. On the other hand, the leach residues from electrolytic plants can contain elevated cadmium levels and care must be exercised in the dumping of such wastes. A novel process, developed by Metallgesellschaft and adopted by the Datteln plant in the F.R.G., avoids the disposal of such large quantities of cadmium to landfills.

### *Primary zinc plants in the European Community*

At present, there are 18 primary zinc plants operational within the European Community. Of these, only five are essentially thermal facilities, four of which utilize the Imperial Smelting Process, and one the vertical retort system. Appendix 1 lists the names, locations and types of smelters in the EC. The production capacity figures for zinc are also provided, as are recent production data, where available.

### *Atmospheric emission estimates*

A request for data on atmospheric cadmium emissions from the five thermal zinc plants was made at the outset of the present study. Three plants, Duisburg, Noyelles Godault, and Avonmouth, provided estimates of an emission factor ( $\text{g Cd t}^{-1} \text{ Zn produced}$ ) and the total quantity of cadmium emitted to the atmosphere in 1979. The relevant values are shown in Table 1. No emission information was available from the ISF smelter at Porto Vesme, Sardinia, or the vertical retort at Harligerode. The emission estimates for these two plants, given in Table 1, were therefore made independently of the companies concerned. For this reason, the assigned values must be viewed with caution.

The low emission value for the Duisburg smelter is related to the absence of any cadmium refining facilities at this plant. Cadmium carbonate produced from sinter fume is refined at the company's electrolytic plant at Datteln (see Appendix 1). Cadmium emission data at the Noyelles Godault plant were available for the main stack only. The

**Table 1 Thermal zinc plants in the European Community: zinc production and estimated airborne cadmium emissions during 1979**

| Location of plant         | Zinc production<br>( $\text{t y}^{-1}$ ) $\times 10^3$ | Cadmium<br>emission<br>factor<br>( $\text{g Cd t}^{-1}$ ) | Cadmium<br>emission<br>( $\text{t y}^{-1}$ ) |
|---------------------------|--------------------------------------------------------|-----------------------------------------------------------|----------------------------------------------|
| Duisburg (F.R.G.)         | 75                                                     | 10-15                                                     | 0.8                                          |
| Noyelles Godault (France) | 86                                                     |                                                           | 5                                            |
| Avonmouth (U.K.)          | 77.4                                                   | 35-52                                                     | 2.7                                          |
| Porto Vesme (Italy)       | 43*                                                    | 70                                                        | 3                                            |
| Harligerode (F.R.G.)      | 81.2                                                   | 100                                                       | 8                                            |

\* Estimated from annual zinc production capacity of the plant and total Italian zinc production in 1979.

company representative suggested that the total emission could be estimated by doubling the main stack value. Additionally, it should be noted that a lead smelter is also present on the site and this presumably contributes to the total emissions.

In view of the relatively small quantities involved, no formal requests for atmospheric emission data were made to the owners of electrolytic refineries. Total atmospheric emissions of cadmium from the electrolytic plant at Datteln, F.R.G., were estimated to be only  $30 \text{ kg y}^{-1}$  (Röpenack 1978). This corresponds to an emission factor of  $0.2 \text{ g Cd t}^{-1}$  zinc produced. Application of this factor to the estimated quantity of zinc produced electrolytically in the EC in 1979 ( $928 \times 10^3 \text{ t}$ ) results in an atmospheric emission of less than  $0.2 \text{ t y}^{-1}$ . The validity of this estimate relies upon the assumption that plant conditions at the other 11 electrolytic installations are similar to those at Datteln. Nevertheless, even if atmospheric emissions are five times greater than estimated, less than  $1 \text{ t y}^{-1}$  cadmium will be released from this source.

#### *Estimates of inputs to water and land*

It is estimated that 50 tonnes of cadmium were released to waters in the EC from zinc production in 1979. Individual facilities vary greatly both in the quantity of cadmium released and also in the characteristics of the water body which receives the effluent. Thus, the ISF plant at Avonmouth is thought to release  $3 \text{ t y}^{-1}$  to the Severn Estuary while the smelter at Duisburg releases about  $1 \text{ t y}^{-1}$  to the River Rhine. The effluent from the electrolytic plant at Datteln also enters the River Rhine, but this is estimated to contain less than  $40 \text{ kg y}^{-1}$ .

The total quantity of cadmium entering landfills from zinc production in 1979 is estimated to be 200 tonnes. Most of this consists of leach residue which is disposed of in landfills constructed with impermeable linings. As pointed out earlier, however, at least one company has adopted a cleaning process which greatly reduces the quantities of cadmium present in solid wastes from electrolytic plants.

#### *1.3.2 Copper and lead*

The production of copper from its ores involves the following operations: (1) roasting of the copper concentrate to remove excess sulphur and produce a matte of copper, iron and some sulphur; (2) conversion of the copper matte to blister copper by fire refining.

At present, production of copper from ore concentrates occurs only in two Member States—Belgium and the Federal Republic of Germany. In 1979,  $159.7 \times 10^3$  t copper were produced from ores. Larger quantities of copper are produced in the EC by the refining of imported copper blister; it has been estimated that  $487 \times 10^3$  t copper originated from this source in the EC in 1979.

The initial roasting of copper concentrates occurs at about  $1200^\circ\text{C}$  and this will result in the vaporization of some of the cadmium present in the concentrates. An EPA study (Deane, Lynn and Suprenant 1976) provided an emission factor of  $3 \text{ g Cd t}^{-1}$  copper produced for primary copper production. No emission data are available for the refining of copper blister but the use of a "cleaner" starting material and the elimination of a roasting stage is expected to result in a lower emission of cadmium. For this reason it has been assumed that the atmospheric emission factor for the refining of copper blister is half that given for the production from ores. Application of these emission factors to the 1979 production data given above, results in an estimated atmospheric emission of about 1.5 t.

Secondary production of copper is a rather general term and includes the smelter production of copper from scrap and waste, the secondary production of refined copper and the direct use of scrap. Secondary production of copper from scrap involves three operations which result in the emission of cadmium to the atmosphere: (1) the melting of the scrap in a blast furnace; (2) the oxidation of impurities in the scrap in a converter; (3) the refining of the copper, typically in a reverberatory furnace.

Experimental studies of several scrap refineries in the U.K. indicate that, of the three stages, converting is responsible for the largest emission of cadmium. This investigation also estimated that total atmospheric emissions of cadmium were unlikely to exceed  $4 \text{ g Cd t}^{-1}$  copper produced. It should be stressed, however, that the quantity of cadmium emitted from such facilities will be strongly influenced by the amount of cadmium present in the scrap. Indeed, the emission factor for those refineries handling copper-cadmium alloy scrap is likely to be much greater than  $4 \text{ g t}^{-1}$ .

In 1976, the last year for which complete figures are available, secondary copper production totalled  $1,132.2 \times 10^3$  t (CEC 1979). This value was made up of  $88.3 \times 10^3$  t of smelter production from scrap and waste,  $326.2 \times 10^3$  t secondary refined copper production and  $717.7 \times 10^3$  t from

direct use of scrap. Application of the emission factor of  $4 \text{ g t}^{-1}$  to total secondary copper production results in an estimated atmospheric cadmium emission of 4.5 t.

Total atmospheric cadmium emissions from primary and secondary copper production have thus been estimated at  $6 \text{ t y}^{-1}$ .

The lack of any published information makes any estimate of cadmium inputs to land and water from copper production rather tentative. In fact, the only estimate available for the EC was provided by Van Wambeke (1979) who gave a figure of 15 t cadmium discharge to landfills from "copper metallurgy" in 1975.

Primary lead production in EC countries occurs by two major routes: the blast furnace smelting of sulphide ores and the ISF technique, where lead is co-produced with zinc. Both processes produce an impure lead bullion which requires further refining. In 1979, the total production of refined lead in the EC amounted to  $1,244 \times 10^3 \text{ t}$  (CEC 1979). About one-third of this quantity was actually produced from secondary materials, the most important being discarded lead-acid batteries. Indeed, many "primary" lead plants are fed with a mixed charge of roasted ores and scrap materials. It has thus been assumed that of the  $1,244 \times 10^3 \text{ t}$  of refined lead produced,  $829 \times 10^3 \text{ t}$  was derived from primary sources and  $415 \times 10^3 \text{ t}$  from secondary materials. The quantity of lead recovered from scrap in 1979 totalled  $837 \times 10^3 \text{ t}$  and this should be added to the secondary refined lead production value, to give the estimated overall secondary lead production figure ( $1,252 \times 10^3 \text{ t}$ ).

At present, there are no published investigations in the EC of cadmium emissions arising from the production of lead. An American survey (Coleman *et al.* 1979) assigned an atmospheric emission factor of  $2.5 \text{ g t}^{-1}$  to primary lead production. However, in an experimental study (Dugdale and Hummel 1978), a primary lead smelter in New Brunswick was estimated to emit more than  $3 \text{ t Cd y}^{-1}$ , which is equivalent to an emission factor of about  $55 \text{ g t}^{-1}$ . It is not clear at present whether this value is representative of primary lead smelters in general. For the present, an emission factor of  $5 \text{ g t}^{-1}$  has been assumed from this source. Application of this factor to the estimated 1979 primary lead production value results in an atmospheric emission estimate of  $4 \text{ t Cd y}^{-1}$ .

A large proportion of the material used for secondary lead production consists of relatively pure scrap lead. For this reason, the emission factor of cadmium from this source will be lower than from primary lead production and has been assumed to be half the latter value, at

2.5 g Cd t<sup>-1</sup>. It is thus estimated that secondary lead production in the EC resulted in an atmospheric cadmium emission of 3 t in 1979.

Total atmospheric cadmium emissions from lead production were assumed to amount to 7 t in 1979.

In view of the paucity of available information, any estimate of the inputs of cadmium to water and land from lead production must at present be tentative. The New Brunswick lead smelter, discussed earlier, was estimated to discharge 3 t Cd y<sup>-1</sup> in the effluent, while the furnace slag contained 7 t Cd y<sup>-1</sup> (Dugdale and Hummel 1978). It is felt that these quantities, and the corresponding emission factors, are exceptionally high and not applicable to lead production in the EC. For the present, it is assumed that lead production in 1979 resulted in cadmium inputs of 20 t to water bodies in the EC and 40 t to landfills.

### *1.3.3 Future trends*

Since 1970, there has been an annual increase in zinc production in emissions from non-ferrous metal production of about 2.5 per cent in the EC. It is considered that this growth will not be maintained in the future and a recent expert study (CEC 1979) considers that the rate will lie between 1.5 and 2 per cent. Taking an average growth rate of 1.7 per cent, it has been calculated that zinc production in 1990 will total  $1,553 \times 10^3$  t and in 2000,  $1,838 \times 10^3$  t. At present, zinc production capacity in the EC is about  $1,600 \times 10^3$  t, but by 1985 it has been estimated that total capacity will be  $1,916 \times 10^3$  t (CEC 1979). Thus, in theory, existing operational plants will be able to accommodate the increased demand for zinc over the next 20 years.

Predicted emissions of cadmium from future zinc production have been estimated after making the following assumptions:

1. Thermal and electrolytic processes will maintain the same share of the zinc market as they do at present.
2. Emission factors to environmental compartments will be the same as at present.

Total atmospheric emissions of cadmium (essentially from thermal plants) are forecast to be 25 t in 1990 and 28 t by 2000. The quantity of cadmium entering landfills, largely from electrolytic facilities, is estimated to be 240 t in 1990 and 286 t in 2000. Waterbodies are predicted to receive a total of 60 t cadmium from zinc production in 1990 and 70 t in 2000.

It must be stressed that the above forecasts can only be regarded as tentative estimates. These values rely on some simplistic economic assumptions as well as the extrapolation of unpublished emission data from individual plants to other facilities. To illustrate the uncertainty which should be attached to these predictions, the only vertical retort in the EC, at Harlingerode, F.R.G. is, according to the owners, shortly to become a secondary plant. If this occurs, then the atmospheric emissions from this plant are expected to diminish. Additionally, the installation of more efficient control devices at certain of the zinc plants in the EC may take place in the near future. This action may reduce cadmium inputs to one environmental compartment but increase inputs to another.

A recent survey of the copper industry, sponsored by the Commission of the European Communities (CEC 1979) has reviewed several forecasts of future demand for copper, both in Europe and the rest of the world. The report concluded that the growth rate in the EC will diminish in the future, and predicted an annual growth rate of 1.7 per cent. For the purposes of this study, it has been assumed that this rate will also occur in the primary and secondary production of copper in the region. Application of this figure to the 1979 copper production data results in a predicted copper production of  $2,142 \times 10^3$  t in 1990 and  $2,535 \times 10^3$  t by 2000. Assuming that the three sources of copper production will maintain their present share of the market (ore production 9 per cent, copper blister 24 per cent, and secondary production 64 per cent), it has been estimated that atmospheric emissions of cadmium from this source will total 7 t in 1990 and 8 t in 2000.

The quantity of cadmium entering landfills from the copper industry has been tentatively estimated to be 20 t by 1990 and 25 t by 2000.

On the basis of a recent expert study (CEC 1979), it is estimated that lead production will increase at an annual rate of 1 per cent in the EC. Application of this value to the total lead production in 1979 ( $2,081 \times 10^3$  t) results in a predicted production of  $2,323 \times 10^3$  t in 1990 and  $2,566 \times 10^3$  t by 2000. Assuming that secondary lead production will, as at present, contribute 60 per cent of the total lead produced, it is predicted that atmospheric cadmium emissions will be 8 t in 1990 and 9 t in 2000. The quantity of cadmium entering landfills has been estimated at 45 t in 1990 and 49 t by 2000. Water bodies in the EC are forecast to receive 22 t cadmium in 1990 and 25 t by the year 2000.

## 1.4 The mining of non-ferrous metals

Non-ferrous metal mines, particularly those which exploit lead-zinc ore fields, are considered to be a significant source of environmental cadmium. At present, however, insufficient information is available to estimate the magnitude of cadmium discharges from mines in the EC. The cadmium content of the ore body, mining management practice, the topography of mine site and the proximity of adjacent watercourses all influence the quantities of cadmium lost from individual sites.

Both wind-blown dispersal of spoil heap material, as well as the activities involved in the processing of extracted ores cause airborne contamination of cadmium from mines. The size of this atmospheric input is unlikely to be large and is expected to result only in localized dispersal of cadmium.

Waterborne dispersal is the major route of cadmium loss from non-ferrous metal mines. Cadmium transport by mine drainage waters can result in the contamination of agricultural soils some distance downstream of the ore field. Flood and storm conditions will increase the mobilization of particulate matter into local watercourses. This problem is not confined to active mines: inadequate management of disused lead-zinc mines has resulted in a continuing contamination of adjacent water bodies (Johnson and Eaton 1980).

Active lead-zinc mines are currently found in four Member States, Italy, F.R.G., France and the Republic of Ireland. No published discharge data are available from these mines despite the probable importance of waterborne cadmium contamination. Van Wambeke (1979) assigned a value of 60–150 t Cd y<sup>-1</sup> lost to waters from active and disused lead-zinc mines in the EC. The basis of this estimate was, however, not stated.

### 1.4.1 *Future trends*

Persistent waterborne dispersal of heavy metals from derelict lead-zinc mines can be controlled by suitable environmental management techniques. Cadmium discharges from these sites could therefore be reduced in the future.

Some of the active lead-zinc mines in the EC have only recently become operational. The largest mine in France, St. Salvay, started zinc concentrate production in 1976, while the Navan mine in the Republic of Ireland only became operational in 1977. Expansion of these mines

is expected in the future and may result in increasing waterborne cadmium discharges.

### 1.5 Production and disposal of cadmium-containing materials

At present, there is insufficient information available to allow an accurate estimation of cadmium discharges from the manufacture and disposal of cadmium-containing materials in the EC. A previous analysis of the situation has been attempted (Rauhut 1978) but the basis of the estimated values is not clear. For this reason, the assumptions given below are derived from the findings of an experimental investigation into the cadmium industry in the U.S.A. (Yost 1979). The estimated discharges provided must therefore be considered provisional and may require significant alteration when relevant data from the EC become available.

It should be stressed that many cadmium-containing products have a long lifetime. For the sake of simplicity, this factor has been ignored when estimating the environmental fate of these products. Additionally, the losses of cadmium which occur during product use have not been taken into account.

#### *Electroplating*

Table 2 shows the estimated fate of cadmium associated with the manufacture of electroplated items for each Member State of the EC. The following assumptions were made in calculating the flow pattern depicted:

1. Of the cadmium consumed in the production of plated items 10 per cent is lost to aqueous wastes and the remaining 90 per cent is present on the product.
2. Of the cadmium in aqueous wastes, 50 per cent is recovered as residues and 50 per cent is discharged to sewage treatment plants ("water").
3. The residues recovered are not reprocessed and are disposed of in landfills.
4. Of the cadmium present in the product, 40 per cent enters the scrap steel cycle and 60 per cent is landfilled.

**Table 2 Fate of cadmium associated with electroplating manufacture and disposal in the EC**

| Country         | Cadmium consumption*<br>(tonnes) | Compartmental flow of cadmium (tonnes) |       |                   |
|-----------------|----------------------------------|----------------------------------------|-------|-------------------|
|                 |                                  | Landfill                               | Water | Scrap steel cycle |
| Italy           | 118                              | 70                                     | 6     | 42.5              |
| The Netherlands | 5                                | 3                                      | .3    | 1.8               |
| U.K.            | 369                              | 218                                    | 18    | 132.7             |
| F.R.G.          | 400                              | 236                                    | 20    | 143.8             |
| Belgium         | 40                               | 24                                     | 2     | 14.4              |
| France          | 275                              | 162                                    | 14    | 98.9              |
| EC Total        | 1207                             | 713                                    | 60    | 434               |

\* Consumption in 1975. Source: Hiscock (1978).

### *Pigments, stabilizers, nickel-cadmium batteries and alloys*

The following assumptions were made in estimating the flow of cadmium from the production and disposal of the other major uses of cadmium:

#### *Pigments*

1. No significant atmospheric emissions of cadmium occur in the manufacture of pigments.
2. All solid residues produced during pigment manufacture are recovered and recycled.
3. Of the cadmium used in pigment manufacture, 2 per cent is ultimately discharged to sewage treatment plants ("water").
4. All the cadmium present in the product ultimately enters the waste disposal pathway.

#### *Stabilizers*

1. Of the cadmium consumed, 50 per cent is used for liquid stabilizer production and 50 per cent for solid stabilizers.
2. Of the cadmium used in the manufacture of liquid stabilizers, 2 per cent is present in filter residues and this is disposed of in landfills.
3. Of the cadmium consumed in the production of solid stabilizers, 2 per cent is not reclaimed from aqueous wastes and is lost to sewage treatment plants ("water").
4. All the product ultimately enters the waste disposal pathway.

### *Nickel-cadmium batteries*

1. Of the total cadmium consumed, 50 per cent is used for pocket-plate cells and 50 per cent for sintered-plate or sealed cells.
2. Of the cadmium used to produce pocket-plate cells, 3 per cent is lost as dusts; 10 per cent of the cadmium in these dusts is discharged to sewage treatment plants ("water") and 90 per cent enters landfills.
3. Of the cadmium present in pocket-plate cells, 50 per cent is recycled and the remainder enters the waste disposal pathway.
4. Of the cadmium consumed in the manufacture of sintered-plate and sealed cells, 5 per cent is lost to aqueous wastes.
5. Of the cadmium in aqueous wastes, 45 per cent is recovered and landfilled, while the remainder is discharged to sewage treatment plants ("water").
6. The cadmium present in sintered-plate and sealed cells enters the waste disposal pathway.

### *Alloys*

Insufficient data exist to quantify the flow of cadmium from the manufacture and disposal of cadmium alloys. Nevertheless, losses of cadmium to landfills and water from product manufacture are unlikely to be significant. Additionally, it is considered that atmospheric emissions from the production of copper-cadmium alloys in the EC will be less than  $3 \text{ t Cd y}^{-1}$ .

Disposal of articles containing cadmium alloys will occur via the secondary copper industry and the waste disposal pathway.

Table 3 depicts the fate of cadmium associated with the manufacture and subsequent disposal of pigments, stabilizers, and nickel-cadmium batteries in the EC. Comparison of the values in Tables 2 and 3 reveals that the quantity of cadmium discharged to water from electroplating units is greater than the combined total from all other major uses. The waste disposal pathway is estimated to receive a total of  $2,643 \text{ t Cd y}^{-1}$  from the disposal of pigments, stabilizers and nickel-cadmium batteries.

#### *1.5.1 Future trends*

There are some signs that cadmium consumption by the plating industry will decline in the future. At present, however, there is little indication of such a reduction in the EC and for this reason it is assumed that

**Table 3 Fate of cadmium associated with the manufacture and disposal of pigments, stabilizers, and nickel-cadmium batteries in the EC**

| Use             | Cadmium consumption* (tonnes) | Compartmental flow of cadmium (t) |               |                |         |
|-----------------|-------------------------------|-----------------------------------|---------------|----------------|---------|
|                 |                               | Landfill                          | Water pathway | Waste disposal | Recycle |
| Pigments        | 1315                          | —                                 | 26            | 1289           |         |
| Stabilizers     | 722                           | 7                                 | 7             | 708            |         |
| Ni-Cd batteries | 900                           | 22                                | 13.7          | 646            | 218     |

\* Consumption data for pigments and stabilizers refer to 1975; those for batteries are the author's estimate for 1980, assuming an annual increase of 5 per cent from 1975 (Source of 1975 data: Hiscock (1978)).

cadmium discharges from this industry will remain constant over the next two decades.

The consumption of cadmium in the manufacture of pigments and stabilizers in the EC is expected to remain stable over the next few years (Hiscock 1978). Discharges of cadmium from these sources are also assumed to remain constant over the next 20 years.

There is a general consensus in the European battery industry that an annual growth of 5 per cent in the production of nickel-cadmium cells will be maintained in the future (Hiscock 1978). This growth rate has been applied over the 20-year period and cadmium consumption by battery manufacture is predicted to be 1,500 t in 1990 and about 2,000 t by 2000. Cadmium discharges have also been assumed to increase at the same rate as future consumption for this industry.

### 1.6 The iron and steel industries

Most of the cadmium released during the production of iron and steel is derived from the recycling of galvanized and cadmium-plated steel scrap. Additionally, the primary raw materials of the steel industry can also contain significant amounts of cadmium and this has to be taken into account when estimating the quantities of cadmium released from this source.

The environmental flow of cadmium through the iron and steel industries has been the subject of a recent study in the U.K. This investigation, detailed in Appendix 2, has provided much of the data base in terms

**Table 4 Cadmium emission factors for the European iron and steel industries, derived from the U.K. study**

| Process                  | Emission factor, g Cd t <sup>-1</sup> material produced |              |       |
|--------------------------|---------------------------------------------------------|--------------|-------|
|                          | Atmosphere                                              | Solid wastes | Slags |
| Sinter production        | 0.08                                                    | 0            | 0     |
| Iron production          | 0                                                       | 0.67         | ND    |
| Basic oxygen steelmaking | 0.02                                                    | 0.36         | 1.11  |
| Electric arc steelmaking | 0.40                                                    | 3.65         | 0.29  |

ND—Not determined.

of emission factors for the estimation of the quantities of cadmium released by other Member States. Table 4 lists the emission factors to air, solid wastes and slags derived from this U.K. study. The term "solid wastes" refers to the flue dusts arrested by collection devices, while slag is the material which remains in the furnace. The higher atmospheric emission factor for electric arc steelmaking (EAS) compared with basic oxygen steelmaking (BOS) reflects the differences in the charge materials used in the two processes. Electric arc furnaces generally receive a charge consisting entirely of steel scrap, while oxygen furnaces have a low scrap loading, the charge consisting mainly of iron. Large quantities of coke are produced for use in the iron and steel industries but the release of cadmium from this process is considered separately, in Section 1.7. Additionally, the amount of cadmium discharged in aqueous wastes from the steel industry has not been estimated, as it is considered that at present there are insufficient data to justify such a calculation.

The experimental study, discussed above, did not consider open-hearth steelmaking as this process was discontinued in the U.K. at the end of 1979. However, some Member States still retain open-hearth plants and an atmospheric emission factor of 1 g t<sup>-1</sup> has been adopted for this process, based on the study of Rauhut (1978). Data on the presence of cadmium in slags and solid wastes were not available for open-hearth steelmaking, so no estimates of the quantities discharged to these compartments could be made.

Table 5 shows the estimated quantities of cadmium discharged by the EC iron and steel industries in 1979, both for individual countries and for processes. A more complete breakdown of these discharges is

**Table 5 Estimated discharges (in tonnes) of cadmium from the EC iron and steel industries in 1979**

(a) for individual Member States

(b) according to process

|                             | Discharge medium |              |       |
|-----------------------------|------------------|--------------|-------|
|                             | Air              | Solid wastes | Slags |
| <b>(a) Country</b>          |                  |              |       |
| Italy                       | 7.7              | 58.3         | 15    |
| Luxembourg                  | 0.7              | 4.4          | 5.5   |
| The Netherlands             | 0.9              | 5.2          | 6.1   |
| United Kingdom              | 5.9              | 40           | 16.4  |
| Federal Republic of Germany | 10.7             | 59.5         | 40.9  |
| Belgium                     | 1.4              | 14           | 14.5  |
| France                      | 6.3              | 46           | 21.7  |
| Denmark                     | 0.5              | 1.5          | 0.1   |
| Total                       | 34               | 229          | 120   |
| <b>(b) Process</b>          |                  |              |       |
| Ironmaking                  |                  |              |       |
| Sinter production           | 10.6             | 0            | 0     |
| Pig iron                    | 0                | 65.8         | ND    |
|                             | 10.6             | 65.8         |       |
| Steelmaking                 |                  |              |       |
| BOS                         | 2.1              | 36           | 111   |
| EAS                         | 13.9             | 128          | 9     |
| OHS                         | 7.6              | ND           | ND    |
|                             | 23.6             | 164          | 120   |

ND—Not determined.

BOS—Basic oxygen steelmaking.

EAS—Electric arc steelmaking.

OHS—Open-hearth steelmaking.

provided in Appendix 3. An additional "sink" for cadmium is the amount retained in the steel itself; applying the U.K. findings to 1979 EC production ( $139 \times 10^6$  t) indicates a total of about 70 t cadmium were present in crude steel.

Atmospheric emissions from sinter and steel production for The Netherlands were not estimated from the emission factors given in Table 4. In this case, the values were obtained from a survey of cadmium in the EC iron and steelmaking industries (C. Steiner, pers. comm.). Other data, provided by the French iron and steel industries, suggest that some of the emission factors in Table 4 may be inappropriate for France. This country relies on domestic ore which is relatively low in iron content. Consequently, larger quantities of ore have to be processed in order to obtain the same tonnage of iron. Furthermore, iron ore from the Lorraine region is reported to contain  $10 \mu\text{g g}^{-1}$  cadmium, compared with only  $0.4 \mu\text{g g}^{-1}$  for ore used in the U.K. It would thus appear that cadmium discharges from iron production in France may be greater than those given in Appendix 3. In the case of electric arc steelmaking, the arrested dusts (solid wastes) from French furnaces have a cadmium content of about  $600 \mu\text{g g}^{-1}$ , compared with  $300 \mu\text{g g}^{-1}$  for U.K. dusts. For this reason, the discharge factors applied to atmospheric emissions and solid wastes for France are double those given in Table 4. Further justification for this assumption can be obtained by estimating the quantity of cadmium entering electric arc steelmaking from steel scrap. In 1978,  $7.5 \times 10^6$  t scrap were consumed by steel production in France, of which 95 per cent was used by electric arc steelmaking (International Iron and Steel Institute, pers. comm.). Assuming that this scrap contained the same cadmium concentration as U.K. scrap ( $\sim 5.5 \mu\text{g g}^{-1}$ —see Appendix 3) then 39 t cadmium would have entered electric arc plants. Application of the standard discharge factors to electric arc production would result in 22 t cadmium unaccounted for, but when the “doubled” discharge factors are used, only 7 t remain outstanding.

Examination of the values given in Table 5(a) reveals that the iron and steel industries in Luxembourg, The Netherlands and Belgium produce relatively low atmospheric emissions of cadmium. This finding is related to both the modest production rates of these countries and to the fact that almost all the steel is produced by the basic oxygen process. The higher atmospheric emission estimates for Italy and U.K. are a consequence of the larger quantities of steel produced, together with the reliance placed upon electric arc steelmaking by these countries.

The Federal Republic of Germany is the largest producer of iron and steel in the EC and was also estimated to emit more cadmium to the atmosphere than the other Member States. Inspection of these emissions by process (Appendix 3) indicates that, in contrast to Italy and the U.K.,

the largest source of cadmium is open-hearth steelmaking followed by sinter production. The relatively small contribution from electric arc steelmaking reflects its minor share in the steel market of F.R.G.

Atmospheric inputs of cadmium from the German iron and steel industries are estimated to total 10.7 t; this figure is considerably higher than the 6 t provided by the German Iron and Steel Federation (C. Steiner, pers. comm.). The present study's choice of Rauhut's atmospheric emission factor for open-hearth steelmaking may be the source of this disparity, as this value is nearly 20 times higher than that given for this source by an EPA study (Coleman *et al.* 1979). There is thus some uncertainty attached to the emissions estimate for this form of steelmaking and experimental work is required to clarify the importance of this source of atmospheric cadmium.

The quantity of cadmium present in steel slags is estimated to be 120 t; this value is similar to the amount of cadmium entering landfills from zinc production in the EC. It should be made clear, however, that the cadmium present in steel slags is associated with much larger quantities of material. Furthermore, the cadmium in these slags is incorporated into a fused mass of material and is thus likely to display a low solubility. Slags from steel production are either disposed of in landfills or utilized commercially for such purposes as road building and the manufacture of concrete.

The solid wastes derived from steelmaking are generally not recycled through the industry and have to be disposed of in landfills. Thus, it is estimated that over 200 t cadmium entered landfills from iron and steel production in the EC. Cadmium in solid wastes is present at higher concentrations than in slags and is associated with fine particulate matter. These factors suggest that this cadmium will display a greater potential for leaching than the cadmium in steel slags. This point is particularly important for electric arc steelmaking, where the cadmium concentrations of the solid wastes are the highest of the three major steelmaking processes (Appendix 3; Yost 1979):

#### 1.6.1 Future trends

Great difficulties exist in making long-term forecasts for iron and steel production in the EC. Most experts in the industry are unwilling to commit themselves to predictions of more than five years in the future. For the purposes of this study it has been assumed that iron and steel production will increase at an annual rate of two per cent in the EC,

with each country maintaining its share of total production. Additionally, it has been assumed that open-hearth and basic Bessemer steelmaking will be phased out by 1990 and their contribution to production replaced by electric arc steelmaking. This assumption can be justified as it is

**Table 6 Summary of predicted discharges (in tonnes) of cadmium from the EC iron and steel industries for 1990 and 2000**  
(a) for individual Member States  
(b) according to process

|                             | Cadmium in discharge medium, 1990/2000 |              |             |
|-----------------------------|----------------------------------------|--------------|-------------|
|                             | Air                                    | Solid wastes | Slags       |
| <b>(a) Country</b>          |                                        |              |             |
| Italy                       | 8.7/10.6                               | 77.4/94.0    | 19.1/23.2   |
| Luxembourg                  | 0.8/1.1                                | 5.3/6.6      | 6.9/8.3     |
| The Netherlands             | 1.2/1.4                                | 6.5/7.9      | 7.6/9.2     |
| United Kingdom              | 6.3/7.7                                | 55.5/67.1    | 20.9/25.5   |
| Federal Republic of Germany | 10.0/12.1                              | 94.6/115.8   | 52.0/63.3   |
| Belgium                     | 1.7/2.2                                | 17.4/21.3    | 18.0/22.0   |
| France                      | 8.5/10.4                               | 66.8/81.7    | 27.5/33.6   |
| Denmark                     | 0.4/0.4                                | 3.3/3.7      | 0.3/0.3     |
| Total                       | 37.6/45.9                              | 326.8/398    | 152.3/185.4 |
| <b>(b) Process</b>          |                                        |              |             |
| <b>Ironmaking</b>           |                                        |              |             |
| Sinter production           | 13.2/16.0                              | 0            | 0           |
| Pig iron                    | 0                                      | 81.3/99.9    | ND          |
|                             | 13.2/16.0                              | 81.3/99.9    |             |
| <b>Steelmaking</b>          |                                        |              |             |
| BOS                         | 2.4/2.98                               | 44.8/54.4    | 137.9/167.9 |
| EAS                         | 22.0/26.9                              | 200.7/243.8  | 14.1/17.2   |
|                             | 24.4/29.9                              | 245.5/298.2  | 152.0/185.1 |

ND—Not determined.

BOS—Basic oxygen steelmaking.

EAS—Electric arc steelmaking.

generally considered that small electric arc furnaces will become increasingly important in the future.

Predicted future production levels and the corresponding discharges are provided in Appendix 4. Those emission factors used for estimating present-day discharges were employed to estimate these predicted discharges. Table 6 summarizes the predicted discharges for individual Member States and for each process.

The most notable change in predicted discharges compared with those of the present day is the increased quantity of cadmium associated with solid wastes. By 2000, it is forecast that about 400 t cadmium will be present in solid wastes, a value almost double the 1979 estimate. It is conceivable that the large quantities of solid wastes with elevated cadmium levels produced by Italy, U.K., F.R.G., and France will pose problems of disposal for these countries.

Atmospheric emissions of cadmium for 1990 are similar to those estimated for the present day. This is a direct consequence of assuming that the open-hearth processes will be discontinued in the near future. By 2000, however, atmospheric emissions are forecast to increase significantly compared with present-day estimates.

## 1.7 Fuel combustion

### 1.7.1 Coal combustion

Cadmium, like many other trace elements, is a natural constituent of coal. Table 7 summarizes the available data on cadmium levels in coal

**Table 7 Cadmium content ( $\mu\text{g g}^{-1}$ ) of coals sampled from the Federal Republic of Germany and United Kingdom**

| Country of origin<br>and coal type            | Mean | Range   | Sample<br>size | Source                          |
|-----------------------------------------------|------|---------|----------------|---------------------------------|
| F.R.G.                                        |      |         |                |                                 |
| Lignite                                       | 0.02 | —       | 1              | Heinrichs (1977)                |
| Bituminous coals                              | 2.0  | 1-10    | 27             | Kautz <i>et al.</i> (1975)      |
| U.K.                                          |      |         |                |                                 |
| Anthracite and bituminous<br>coals from Wales | 1.0  | 0.4-10  | 302            | Chatterjee and<br>Pooley (1977) |
| "Power station" coals                         | 0.4  | 0.3-0.8 | NG             | Gibson (1979)                   |

NG—Not given.

samples from the F.R.G. and U.K., the two largest coal consumers in the EC. Apparently hard coals from Germany have the higher cadmium content, but values from both countries display a wide range of variation. The mean values given in Table 7 have been taken into account when assigning an average cadmium content of  $1 \mu\text{g g}^{-1}$  to hard coals consumed in the EC. German lignites have also been assumed to contain  $1 \mu\text{g Cd g}^{-1}$ , despite the very low level shown in Table 7. This value was based on a single sample of lignite and may not be representative of values in lignite.

At present, the largest coal-consuming sectors of the EC are electrical power stations and coke ovens; these accounted for 56 per cent and 29 per cent of the total EC production in 1978. Other industries used about 6 per cent while the domestic and commercial sector burnt 7 per cent of the total consumed. The F.R.G. accounts for nearly all the lignite consumed in the EC and about 90 per cent of this is burnt at power stations.

In the past five years, there have been several investigations into the flow of cadmium and other trace elements through coal-consuming facilities but these have been confined to modern pulverized coal power plants with sophisticated particulate control devices. Coal combustion in these facilities occurs at about  $1,500^{\circ}\text{C}$  and results in the formation of a slag or bottom ash which is retained in the furnace. The remainder of the non-combustible mineral matter leaves the furnace as fly ash and is accompanied by the flue gases and volatilized minerals. This flue stream next passes through the particulate control devices where up to 99 per cent of the coarser fly ash particles are removed, but smaller ( $< 1 \mu\text{m}$ ) particles are less efficiently retained. The cadmium content of these waste streams has been found to increase in a consistent manner, in the order bottom ash  $<$  inlet fly ash  $<$  outlet fly ash. This partitioning indicates that cadmium is volatilized when the coal is burnt, but later, when the flue gases cool, it condenses in a preferential manner on the smaller fly ash particles. The selective retention of the coarser fly ash particles by the collection devices will further enhance cadmium's association with submicron particles.

Despite cadmium's small particle association, an emission factor of only  $0.02 \text{ g Cd t}^{-1}$  has been assumed for the combustion of coal in power stations. This figure is based upon the data of Coleman *et al.* (1979), Yost (1979) and Nriagu (1979) and reflects the efficiency of the collection devices used in these facilities.

Little information exists on the emissions of cadmium from the other coal-consuming sectors. For this reason an emission factor of  $0.02 \text{ g Cd t}^{-1}$  has also been assigned to these sectors. In the case of coke production, there is some experimental evidence to support this assumption, as cadmium emissions from coke ovens have been measured as part of a wider study of cadmium in the U.K. steel industry (Appendix 2). The emission estimate of  $0.15 \text{ t}$  cadmium from  $10^7 \text{ t}$  coal corresponds to an emission factor of just under  $0.02 \text{ g t}^{-1}$ .

Apparently, no studies of cadmium emissions from coal burning in domestic appliances have been carried out. Particulate control devices are normally absent from such units, suggesting that emissions will be greater than from power plants on a unit basis. To counter this, the combustion temperature in domestic units is lower than in power plants and this possibly results in a less efficient vaporization of cadmium. In view of the uncertainty attached to the emission characteristics of these appliances, the emission factor assigned to coal combustion in power plants has also been used for domestic units.

Table 8 summarizes the estimated discharges of cadmium from coal consumption in the EC Member States for 1978, the last year for which data are available. Atmospheric emissions total just under  $6 \text{ t}$ , of which

**Table 8 Estimated discharges of cadmium from coal consumption in the EC in 1978**

| Country             | Coal consumption*<br>1978 ( $\times 10^6 \text{ t}$ ) | Cadmium in discharge medium (tonnes) |       |
|---------------------|-------------------------------------------------------|--------------------------------------|-------|
|                     |                                                       | Atmospheric emissions                | Ashes |
| Italy               | 12.1                                                  | 0.2                                  | 12    |
| Luxembourg          | 0.5                                                   | —                                    | 0.5   |
| The Netherlands     | 5.0                                                   | 0.1                                  | 5     |
| U.K.                | 120.6                                                 | 2.4                                  | 118   |
| F.R.G.              | 81.9                                                  | 1.6                                  | 80    |
| Belgium             | 16.6                                                  | 0.3                                  | 16    |
| France              | 44.2                                                  | 0.9                                  | 43    |
| Denmark             | 5.6                                                   | 0.1                                  | 5.5   |
| Republic of Ireland | 0.8                                                   | —                                    | 0.8   |
| EC Total            | 287.4                                                 | 5.8                                  | 282.5 |

\* Source: Eurostat (1980).

the U.K. accounts for about 2.5 t and the F.R.G. 1.5 t. In the case of solid wastes, it is estimated that nearly 300 t of cadmium are present in the ashes retained by the particulate control devices employed in the various coal burning facilities. Fly ash represents the major solid waste from pulverized coal power plants: most of this is disposed of in landfills and ash ponds. The remainder is utilized on a commercial basis for road building and the production of construction materials.

In 1978, power plants in the F.R.G. burnt  $1.1 \times 10^8$  t lignite (Eurostat 1980). The quantity consumed is estimated to result in 2.2 t cadmium released to the atmosphere and 108 t incorporated in ashes.

### 1.7.2 Future trends

In the near future, coal is expected to assume an increasingly important rôle as an energy source, to supplement the decreasing stocks of petroleum and natural gas. The conversion of coal to synthetic fluid fuels is also predicted to become commercially significant as a source of coal consumption in the near future. A brief assessment of the potential cadmium discharges from such technologies is provided in Section 1.7.3. In contrast to hard coal, lignite consumption is not expected to increase markedly over the next two decades.

Forecasts of EC coal and lignite consumption for 1990 and 2000 together with the predicted cadmium discharges are shown in Table 9. Insufficient information was available to allow the estimation to be made on a Member State basis.

**Table 9 Predicted discharges of cadmium from coal and lignite consumption in the EC for 1990 and 2000**

| Year | Predicted consumption<br>( $\times 10^6$ t) | Cadmium in discharge medium (tonnes) |       |
|------|---------------------------------------------|--------------------------------------|-------|
|      |                                             | Atmospheric emissions                | Ashes |
| 1990 | Coal <sup>a</sup>                           | 315                                  | 6.3   |
|      | Lignite <sup>b</sup>                        | 140                                  | 2.8   |
| 2000 | Coal                                        | 530                                  | 10.6  |
|      | Lignite                                     | 150                                  | 3     |

<sup>a</sup> Source of consumption prediction—Official Journal of the European Communities (1979).

<sup>b</sup> Author's own prediction.

### 1.7.3 Coal conversion

Apparently, no investigations have been performed on trace element discharges from the few commercially sized coal conversion plants in existence at present. For this reason, an emissions assessment of bench-scale or pilot-plant versions of three gasification processes and two liquefaction processes has been performed. The five processes considered were selected for their future commercial importance and for the range of reaction conditions and effluent characteristics. The three gasification processes are the synthane gasifier, the Koppers-Totzek gasifier and the Lurgi gasifier. The synthoil and the solvent-refined coal processes were the two liquefaction processes considered. The basic data were obtained from Oldham and Wetherold (1977).

All the processes considered, with the exception of the Koppers-Totzek process, utilize higher pressures and lower temperatures than conventional coal-fired power plants. These conditions indicate that cadmium will be less efficiently volatilized from the feed coal, resulting in a less pronounced partitioning between fly ash and bottom ash and a smaller enrichment on submicron particles. Thus, atmospheric emission factors for coal conversion processes in general are likely to be lower than from coal burning in power plants. For the sake of simplicity, however, the present study has assigned a uniform atmospheric emission factor to future coal consumption (Table 9).

### 1.7.4 Oil and gas

The cadmium content of crude oil is lower than coal (Hofstader, Milner and Runnels 1976) and an average value of  $0.05 \mu\text{g g}^{-1}$  has been assigned to this material. Little information exists on the extent of cadmium release from the combustion of oils. It has been assumed that 2 per cent, the same proportion estimated for coal combustion, will be released when oil is burnt. This corresponds to an emission factor of  $0.001 \text{ g t}^{-1}$ . The majority of the cadmium in oil is thus expected to be retained in ashes.

Fuel oil consumption in the EC totalled  $301 \times 10^6 \text{ t}$  in 1978, resulting in an estimated atmospheric emission of less than 0.5 t cadmium. The remainder of the cadmium in oil, about 14.5 t, will be present in ashes, most of which will be disposed of to landfills or utilized commercially.

Figures for EC oil consumption for 1990 and 2000 have been predicted to be 25 per cent and 20 per cent greater than the 1977 level (CEC

1979). Fuel oil consumption is thus expected to be about  $375 \times 10^6$  in 1990 and  $360 \times 10^6$  t by 2000. The corresponding atmospheric emissions of cadmium are less than 0.5 t for both years. Similarly, solid wastes are estimated to contain about 18 t cadmium for both 1990 and 2000.

Liquid fuels produced by coal conversion are expected to be of commercial importance by the next decade. At present, cadmium emissions from plants burning synthetic oil have not been assessed. However, one particular liquefaction product, solvent refined coal, has been analysed, the cadmium content being  $0.05 \mu\text{g g}^{-1}$  (Oldham and Wetherold 1977). This value is identical to the average level assumed for oil in this study, indicating that the burning of solvent refined coal would also result in similar cadmium emissions.

Natural gas does not contain significant quantities of cadmium and is considered to be a negligible source of cadmium in the environment. Future manufacture of coal gasification products will incorporate sophisticated cleaning procedures to remove all particulate matter. This will, in effect, reduce the cadmium content of such products to zero.

### 1.8 Refuse disposal

The major sources of cadmium in refuse are scrap metal and plastics containing pigments and stabilizers. The heterogeneous composition of refuse presents difficulties when estimating an average cadmium content for this material. Values of  $12 \mu\text{g g}^{-1}$  for the U.K. and  $30 \mu\text{g g}^{-1}$  for the F.R.G. have been reported (DOE 1980); this study has assumed an average cadmium content of  $15 \mu\text{g g}^{-1}$  for the EC.

Table 10 shows the quantities of refuse generated by each Member State in 1977 and the estimated totals disposed of directly in landfills. The large quantities of waste disposed of in this manner result in an estimated 1,152 t cadmium entering such sites. The U.K. and F.R.G. are the two largest contributors and together account for over half this total.

At present, about 20 per cent of the refuse generated in the EC is incinerated. The amounts combusted in 1978 by individual Member States are shown in Table 11. Several studies have reported that fly ash derived from the burning of refuse in waste incinerators contains elevated concentrations of cadmium (Greenberg, Zoller and Gordon 1978, Scott 1979). Furthermore, it has also been shown that cadmium is predominantly associated with submicron particles, the particles least

**Table 10 Quantity of refuse generated in the EC for 1977 and estimated cadmium inputs from direct landfill**

| Country         | Waste generated <sup>a</sup><br>( $\times 10^6$ t) | Waste in landfills <sup>a</sup><br>( $\times 10^6$ t) | Cadmium in landfills<br>(tonnes) |
|-----------------|----------------------------------------------------|-------------------------------------------------------|----------------------------------|
| Italy           | 16                                                 | 13.4                                                  | 201                              |
| Luxembourg      | 0.13                                               | —                                                     | —                                |
| The Netherlands | 5.3                                                | 3.6                                                   | 54                               |
| U.K.            | 28 <sup>b</sup>                                    | 24.8                                                  | 372                              |
| F.R.G.          | 25                                                 | 19.6                                                  | 294                              |
| Belgium         | 3.5                                                | 3                                                     | 45                               |
| France          | 16                                                 | 11.1                                                  | 166                              |
| Denmark         | 2.5                                                | —                                                     | —                                |
| Ireland         | 1.3                                                | 1.3                                                   | 20                               |
| EC Total        | 97.7                                               | 76.8                                                  | 1,152                            |

<sup>a</sup> Source: Van Wambeke (1980).

<sup>b</sup> Source: U.K. Department of the Environment, pers. comm.

**Table 11 Quantity of refuse incinerated in the EC for 1977 and estimated cadmium discharges**

| Country         | Waste incinerated <sup>a</sup><br>( $\times 10^6$ t) | Cadmium in discharge medium (tonnes) |       |
|-----------------|------------------------------------------------------|--------------------------------------|-------|
|                 |                                                      | Atmospheric emissions                | Ashes |
| Italy           | 2.6                                                  | 3.9                                  | 35.1  |
| Luxembourg      | 0.1                                                  | 0.2                                  | 1.4   |
| The Netherlands | 1.7                                                  | 2.6                                  | 23    |
| U.K.            | 3.2 <sup>b</sup>                                     | 4.8                                  | 43    |
| F.R.G.          | 5.4                                                  | 8.1                                  | 72.9  |
| Belgium         | 0.54                                                 | 0.8                                  | 7.3   |
| France          | 4.9                                                  | 7.4                                  | 66    |
| Denmark         | 2.5                                                  | 3.7                                  | 33.8  |
| Ireland         | —                                                    | —                                    | —     |
| EC Total        | 20.9                                                 | 31.4                                 | 282   |

<sup>a</sup> Source: Van Wambeke (1980).

<sup>b</sup> Source: U.K. Department of the Environment, pers. comm.

efficiently retained by particulate control devices. Experimental measurements of trace element emissions from refuse incinerators reveal a relatively high emission factor of cadmium from this source, with values ranging from 0.7–4.4 g Cd t<sup>-1</sup> (Greenberg *et al.* 1978, Scott 1979).

After examining the relevant data available, an emission factor of 1.5 g t<sup>-1</sup> has been assigned to atmospheric cadmium release from refuse incineration. This value is considered to be representative of the larger, modern facilities equipped with sophisticated particulate control devices: however, some of the smaller refuse incinerators do not possess efficient control units and emission rates may be higher than 1.5 g t<sup>-1</sup>.

Atmospheric emissions of cadmium from refuse incineration in the EC were estimated to total over 30 t in 1978 (Table 11). The largest contributors were, in decreasing order, the F.R.G., France and the U.K.

The cadmium retained in the incinerator is partitioned between collected fly ash and the furnace clinker. However, as both wastes are generally landfilled, they have been considered as a single discharge medium in Table 11. The total of 282 t cadmium present in these ashes is considered to be of greater environmental significance than the 1,152 t estimated to be discharged by direct landfill. This is because the cadmium in fly ash is expected to be more soluble than in untreated waste and will thus display a greater propensity for leaching from landfill sites.

The quantity of cadmium associated with waste disposal in the EC is thus estimated to total 1,465 t y<sup>-1</sup>. This value is some 1,000 t less than the amount assumed to enter the waste cycle pathway from the disposal of cadmium-containing products (see Table 2). This disparity may be a result of the earlier assumption that cadmium-containing products are disposed of after only 12 months' use. In reality, such items may have a much longer useful lifetime. Alternatively, it is feasible that the cadmium content of refuse has been underestimated; use of an average value of 30 µg g<sup>-1</sup> in refuse would, for example, produce an annual total of 2,930 t cadmium associated with waste disposal.

### 1.8.1 Future trends

Future cadmium discharges from refuse disposal are difficult to predict. At present, the rate of waste generated in the EC appears to be levelling off (Van Wambeke 1980) but no published forecasts of future patterns are available. The proportion of waste incinerated in the EC may increase in the future, as the rising costs of conventional fuels will make energy production from this source a more attractive proposition. This need

not necessarily result in larger cadmium discharges from this source, because future legislation may restrict the use of the major sources of cadmium in waste material. Controls on the use of cadmium stabilizers and pigments in plastics and cadmium-plated articles would lead to a reduction in the average cadmium content of waste.

In view of these conflicting factors, it has been assumed that cadmium discharges from this source will remain constant over the next two decades.

### 1.9 Sewage sludge

The production of sewage sludge in the EC was estimated by Van Wambeke (1979) to total  $5 \times 10^6$  t (dry weight) in 1975. More recent data have not been published, but it is thought that the total will have risen, as the contributions from some Member States are expected to have increased significantly over the last five years. In the F.R.G. for example,  $1.7 \times 10^6$  t sludge were generated in 1974; by 1985 the quantity has been forecast to be  $4 \times 10^6$  t (Thormann 1979). Other Member States, for example the U.K., are unlikely to display such large increases. For the purposes of this study, therefore, it has been assumed that sludge generation in the EC has increased at an annual rate of 2 per cent, indicating a generation of  $5.5 \times 10^6$  t sludge in 1980.

The cadmium present in sewage sludge originates from both domestic and industrial sources, but the latter is thought to make the major contribution. Cadmium losses to water from the manufacture of cadmium-containing articles in the EC were earlier estimated to total 108 t. If it is assumed that all of this cadmium is discharged to sewers and all retained in sludges, then this source alone would produce a cadmium content of about  $20 \mu\text{g g}^{-1}$  in the estimated  $5.5 \times 10^6$  t sludge produced in 1980. In assigning an average cadmium content to sewage sludge in the EC, the mean value of about  $30 \mu\text{g g}^{-1}$  obtained in an extensive U.K. survey (DOE 1980) has been adopted. Thus, an estimated total of 165 t cadmium are assumed to be present in sewage sludge generated in the EC in 1980.

The pattern of sewage sludge disposal will vary between Member States of the EC but detailed information is only available for three countries, and this is summarized in Table 12. On the basis of these figures, it has been assumed that 75 per cent of the sludge generated in the EC is disposed of via land, 20 per cent in water (sea) and 5 per cent

**Table 12 Sewage sludge disposal patterns for three Member States of the EC**

| Country and authority                             | Sludge generated<br>$\times 10^6$ t (year) | Disposal pathway,<br>percentage of total |      |                |
|---------------------------------------------------|--------------------------------------------|------------------------------------------|------|----------------|
|                                                   |                                            | Inciner-<br>ation                        | Land | Water<br>(sea) |
| The Netherlands<br>Scheltinga (1979)              | 0.137 (1974)                               | 2                                        | 78   | 20             |
| United Kingdom<br>DOE (1980)                      | 1.3 (1979)                                 | 4                                        | 67   | 29             |
| Federal Republic<br>of Germany<br>Thormann (1979) | 1.7 (1974)                                 | 8                                        | 87   | 5*             |

\* Originally assigned under heading "Other processes".

is incinerated. It has been additionally assumed that 25 per cent of the cadmium present in sludge is released to the atmosphere upon incineration and the remainder is ultimately disposed of to land. This high atmospheric emission estimate is based upon the finding that some sewage sludge incinerators in the U.K. release one-third of the cadmium in sludge to air (DOE 1980). Application of this disposal pattern results in an estimated 2 t cadmium emitted to the atmosphere, 33 t to water and 130 t to land.

### 1.9.1 Future trends

The quantity of sewage sludge generated in the EC is expected to continue to increase in the future. Precise forecasts have not been published for most Member States but an annual growth rate of 2 per cent is considered to be a reasonable estimate for the EC as a whole. This growth rate results in a predicted generation of  $6.5 \times 10^6$  t sludge in 1990 and  $8 \times 10^6$  t by 2000. It has additionally been assumed that the estimated average cadmium content of sewage sludge, at present  $30 \mu\text{g g}^{-1}$ , will remain stable over the next two decades. Some uncertainty is attached to this estimate, as cadmium discharges to sewage systems from the manufacture of cadmium-containing articles are predicted to increase only moderately over the next 20 years (Section 1.5.1). Thus, the increased tonnage of sewage sludge generated in the future may not be matched by a comparable rise in cadmium discharges. For the present, however, the value of  $30 \mu\text{g g}^{-1}$  will be used.

The pattern of sewage sludge disposal in the EC is expected to alter in the future. In The Netherlands, for example, sea disposal is expected to decrease while sludge incineration will increase in importance (Scheltinga 1979). The present study has assumed that by 1990, 75 per cent of sludge will be disposed of via land, 15 per cent in water (sea) and 10 per cent incinerated. By 2000, the proportions incinerated and discharged to waters are forecast to be the same, at 15 per cent, while land will receive 70 per cent of the total.

Application of these predicted disposal patterns results in the following discharges of cadmium in 1990 and 2000: atmospheric emissions from sludge incineration 5 and 9 t; discharge to land 161 and 195 t; discharge to waters 29 and 36 t.

## 1.10 Phosphate fertilizers

### 1.10.1 Fertilizer manufacture

The major source of cadmium discharges from phosphate fertilizer manufacture originates from the wet process production of phosphoric acid. This technique uses sulphuric acid to form phosphoric acid and calcium sulphate (gypsum) from rock phosphate. The cadmium originally present in the rock phosphate will become distributed between the phosphoric acid and gypsum waste as a result of sulphuric acid action. The major disposal route of the gypsum is by dumping in coastal waters. A proportion is, however, recovered and utilized by the construction industries.

Estimated discharges of cadmium to land and water from the manufacture of phosphoric acid by the wet process are shown in Table 13. The values have been computed from a recent survey of the phosphate fertilizer industry in the EC (Feenstra 1978). France is the largest phosphoric acid producing country in the EC and also accounts for the major portion ( $27 \text{ t y}^{-1}$ ) of cadmium discharged to waters from this source. In the F.R.G., most of the gypsum is recovered and used as a construction material; as a consequence, cadmium discharges from phosphoric acid manufacture are negligible. The values given in Table 13 are associated with capacity production of wet process phosphoric acid in the Member States. It is unlikely that all, if any, wet process plants in the EC are actually operating at full capacity; the estimated discharges in Table 13 must therefore be considered as maximum values.

**Table 13** Estimated discharges of cadmium from the manufacture of phosphate fertilizers in the EC

| Country                     | Cadmium ( $\text{t y}^{-1}$ ) |       |
|-----------------------------|-------------------------------|-------|
|                             | Landfill                      | Water |
| Italy                       | 1                             | 8.4   |
| The Netherlands             | 1.3                           | 18    |
| United Kingdom              | 20                            | 7.5   |
| Federal Republic of Germany | 0.02                          | 0.05  |
| Belgium/Luxembourg          | 9                             | 0.3   |
| France                      | 2.6                           | 27    |
| Denmark                     | —                             | 0.5   |
| Total                       | 34                            | 62    |

Sources: Feenstra (1978) and DOE (1980).

### 1.10.2 Fertilizer use

The cadmium contents of phosphate fertilizers vary widely and depend upon the country of origin. Table 14 shows the estimated average cadmium levels in fertilizers from the major exporters of rock phosphate to the Member States of the EC. These values were calculated on the basis that no losses of cadmium occur in the production of fertilizer from rock phosphate. This is, of course, not the case for wet process

**Table 14** Assumed cadmium content of phosphate fertilizers according to country of origin

| Country of origin            | Cadmium content <sup>a</sup><br>$\text{g Cd t}^{-1} \text{P}_2\text{O}_5$ |
|------------------------------|---------------------------------------------------------------------------|
| Morocco                      | 60                                                                        |
| U.S.A.                       | 35                                                                        |
| Togo                         | 160                                                                       |
| Senegal                      | 255                                                                       |
| U.S.S.R.                     | 0.8                                                                       |
| Tunisia/Algeria <sup>b</sup> | 60                                                                        |
| Israel/Jordan <sup>b</sup>   | 35                                                                        |

<sup>a</sup> Values modified from Feenstra (1978).

<sup>b</sup> Values assigned by author of present study.

production, but insufficient information exists on the relative importance of this process in the EC to allow a refinement of the figures in Table 14. The quantities of phosphate rock imported by the EC Member States in 1979 have been broken down according to country of origin in Appendix 5. Appendix 5 also gives phosphate fertilizer consumption in the EC for 1979. These data, in conjunction with the values given in Table 14, have been used to estimate the amounts of cadmium discharged to agricultural land in the region as a consequence of fertilizer application. The estimated quantities for each Member State are shown in Table 15. In calculating these discharges, it has been assumed that the origin of fertilizer used in each Member State corresponds to the pattern of rock phosphate importation. Thus, in the case of the U.K., it is assumed that 24 per cent of fertilizer consumed originates from U.S.A. rock phosphate, 24 per cent from Senegal and 52 per cent from Morocco and Tunisia (see Appendix 5).

The total quantity of cadmium discharged to land from fertilizer use in the EC is estimated to be 312 t in 1979.

### 1.10.3 Future trends

Future trends in phosphate fertilizer manufacture and use are difficult to predict. Recent increases in the price of fertilizers may reduce future demand and consumption in the EC. However, this is not expected to reduce significantly the quantity consumed in the long term. Therefore,

**Table 15 Estimated discharges of cadmium to agricultural land from the application of phosphate fertilizers in the EC in 1979**

| Country                     | Total cadmium discharged (t) |
|-----------------------------|------------------------------|
| Italy                       | 49                           |
| The Netherlands             | 7                            |
| United Kingdom              | 45                           |
| Federal Republic of Germany | 44                           |
| Belgium/Luxembourg          | 8                            |
| France                      | 144                          |
| Denmark                     | 7                            |
| Republic of Ireland         | 8                            |
| EC Total                    | 312                          |

for the present it has been assumed that phosphate fertilizer manufacture and consumption will remain constant over the next two decades.

## 1.11 Summary and conclusions

### 1.11.1 Introduction

This section has attempted to quantify the major sources of cadmium inputs into the environment of the EC. The values obtained are summarized in Table 16. In some cases, the assigned values should be considered as tentative until more precise estimates can be made. This is the case where a single cadmium concentration has been assigned to a material known to contain a wide range of concentrations. Additionally, the use of a single emission/discharge factor (g Cd released  $t^{-1}$  material produced/consumed) to a process carried out in facilities with varying degrees of emission control is an oversimplification which may conceal large differences in emissions between different plants.

Cadmium release has been considered for three broad environmental compartments: air, land, water. This approach masks large differences

**Table 16** Summary of current cadmium inputs to the EC environment ( $t\ y^{-1}$ )

| Source                                     | Compartment |       |       |
|--------------------------------------------|-------------|-------|-------|
|                                            | Air         | Land  | Water |
| Volcanic action                            | 20          | ND    | ND    |
| Non-ferrous metal production               |             |       |       |
| Zinc and cadmium                           | 20          | 200   | 50    |
| Copper                                     | 6           | 15    | ND    |
| Lead                                       | 7           | 40    | 20    |
| Production of cadmium-containing materials | 3           | 90    | 108   |
| Iron and steel production                  | 34          | 349   | ND    |
| Fuel combustion                            |             |       |       |
| Coal and lignite                           | 8           | 390   | ND    |
| Oil and gas                                | 0.5         | 14.5  | —     |
| Waste disposal                             | 31          | 1,434 | ND    |
| Sewage sludge disposal                     | 2           | 130   | 33    |
| Phosphate fertilizers                      | —           | 346   | 62    |
| Totals                                     | 132         | 3,009 | 273   |

ND—Not determined.

in the mobility and environmental significance of the cadmium released according to the precise disposal route employed. In the terrestrial compartment, for example, sewage sludge applied to agricultural land will be of greater significance from the perspective of human exposure and environmental mobility than sludge which is landfilled. Similarly, cadmium inputs to the aquatic environment from phosphate fertilizer manufacture and sewage sludge disposal generally occur in coastal waters and are of little direct significance to human exposure. In contrast, aquatic cadmium discharges from electroplating facilities are expected to enter sewage collection systems. A large proportion of the cadmium will be incorporated in the sewage sludge and some of this will subsequently be applied to agricultural land.

Another aspect of importance, not clear from Table 16, is that the environmental significance of cadmium in a particular waste stream is not necessarily directly related to the total quantity of cadmium present. Thus, the 200 t cadmium in solid wastes from zinc production is considered to be of greater potential significance than the 390 t cadmium present in ashes from coal combustion because the former is associated with much smaller amounts of waste material.

#### *1.11.2 The major sources of environmental cadmium in the EC*

The iron and steel industries, together with refuse incineration, represent the two largest sources of atmospheric cadmium in the EC. The other major sources, thermal zinc production and volcanic action, are next in ranking. All other sources account for less than one-quarter of the total atmospheric discharges.

Waste disposal results in the single largest input of cadmium to land in the EC. The quantity of cadmium associated with this pathway is greater than the combined total of the four other major sources. These processes are, in descending rank, coal combustion, iron and steel production, phosphate fertilizer manufacture and use, and zinc production. These five sources together contribute 90 per cent of the total cadmium inputs to land in the region.

As has been pointed out earlier, the characterization of cadmium inputs to aquatic systems in the EC is incomplete. Of the sources considered in the present study, the manufacture of cadmium-containing articles is estimated to result in the single largest input of cadmium. Phosphate fertilizer manufacture is next in ranking, followed by discharges from zinc production.

**Table 17 Cadmium discharges to the environment of the EC: comparison with previously published estimates**

| Authority          | Year      | Cadmium flow ( $\text{t y}^{-1}$ ) |             |         |
|--------------------|-----------|------------------------------------|-------------|---------|
|                    |           | Air                                | Land        | Water   |
| Present study      | 1979/1980 | 132                                | 3,009       | 273     |
| Rauhut (1978)      | 1975      | 263                                | 4,760       | 133     |
| Van Wambeke (1979) | 1975      | 365-435                            | 2,600-2,880 | 217-317 |

### *1.11.3 Comparison of total environmental inputs of cadmium in the EC with previous estimates*

Table 17 depicts the total inputs of cadmium to the three environmental compartments derived from the present study in comparison with two previous estimates for the EC. The estimate provided by the present study for the total atmospheric input of cadmium is much lower than the other values, being half that given by Rauhut (1978) and one-third that assigned by Van Wambeke (1979). The basis for this disparity is considered in Section 1.11.4. Less difference exists in the three estimates for total cadmium discharges to land. However, this similarity conceals large differences in the estimated contributions from individual sources by the three studies. Thus, in Rauhut's study, no account was taken of the cadmium present in solid wastes from either the iron and steel industries or from fossil fuel combustion. Despite this omission, Rauhut's estimate for the total quantity of cadmium discharged to land is still the largest of the three values. In the case of aquatic discharges, the total quantities assigned by the present study and by Van Wambeke are in reasonable agreement. The estimate given by Rauhut is much lower, because only two sources were considered: zinc production and the manufacture of cadmium-containing materials.

### *1.11.4 Comparison with previous inventories of cadmium release according to source*

#### *Zinc production*

In the past, many studies have assumed that zinc production represents the single largest source of atmospheric cadmium. This investigation, however, estimates that zinc production accounts for only 15 per cent of the total discharges to this compartment. The quantity involved,

$20 \text{ t y}^{-1}$ , is similar to the values assumed by Rauhut (1978) and Van Wambeke (1979). This amount is some 10 times smaller than the emission assigned to zinc production in another EC study (Van Enk 1979). Another anomalously high value has recently been given in an EPA study (Coleman *et al.* 1979). This investigation estimated an atmospheric release of  $537 \text{ t y}^{-1}$  in the U.S.A., yet zinc production in the U.S.A. is actually less than in the EC. It is true that zinc production in the U.S.A. relies more heavily on thermal processes but it is felt that an erroneously high emission factor was employed by Coleman *et al.* (1979). The comparison with an American investigation has been made as some workers in Europe point to this high estimate and suggest a similar situation prevails in the EC.

The present study has relied upon unpublished information provided by the relevant companies to estimate emissions from zinc production. Unfortunately, there would appear to be no independent studies of such facilities, in the way that there have been for both coal-fired power plants and refuse incinerators. It is hoped that this situation will be rectified in the future and enable the producers' estimates to be verified.

The quantity of cadmium discharged to water from zinc production was estimated to total  $50 \text{ t y}^{-1}$ ; this figure is about half that given by both Rauhut (1978) and Van Wambeke (1979). Although the value of  $50 \text{ t y}^{-1}$  can only be considered tentative at present, the specific discharge data of three of the 18 zinc plants ( $3 \text{ t y}^{-1}$ ,  $1 \text{ t y}^{-1}$  and  $<40 \text{ kg y}^{-1}$ ) suggest that Rauhut's estimate of  $>100 \text{ t y}^{-1}$  is too large.

Insufficient information was available to make an independent estimate of the quantity of cadmium discharged to land from zinc production. For this reason, the figure of  $200 \text{ t y}^{-1}$  was based in part on the earlier estimate of Rauhut (1978). This quantity is lower than the estimated discharges to land from iron and steel production, coal combustion or waste disposal. Nevertheless, the cadmium concentrations in these residues will be markedly higher than in the solid wastes derived from other sources. Attention should be paid to the possibility of cadmium leaching from landfills containing these residues.

#### *Copper and lead production*

Atmospheric discharges of cadmium from copper production were estimated to be  $6 \text{ t y}^{-1}$ ; this value is similar to the  $5 \text{ t y}^{-1}$  assigned to this source by Van Wambeke (1979). Rauhut (1978) did not take into account cadmium emissions from the copper industry.

It is considered that emissions from secondary copper refineries may have been underestimated in the present study. More information is required to establish whether this is the case. Those facilities processing copper-cadmium alloy scrap require particular attention.

The estimated cadmium discharge to land was based on a value provided by Van Wambeke (1979) and can only be considered as tentative at present. Less stringent precautions may be taken in the disposal of solid wastes from copper production than from zinc production, especially in the case of those refineries which handle cadmium-containing scrap on an irregular basis. At present, however, no studies have been made of the mobilization of cadmium from such landfills.

Cadmium discharges from the lead industry were made on a speculative basis as no appropriate studies have been published on these facilities in the EC. The investigation of a primary lead smelter in the U.S.A. (Dugdale and Hummel 1978) suggests that atmospheric emissions of cadmium from this source may be greater than have been assumed by the present study. Practical investigations of lead smelters in the EC are needed before a more accurate cadmium emissions estimate from this source can be made.

#### *The mining of non-ferrous metals*

The present study has made no attempt to quantify aquatic cadmium discharges from lead-zinc mines in the EC. The value provided by Van Wambeke of about  $100 \text{ t y}^{-1}$  is likely to be a conservative estimate. Discharges from individual mine sites will be very variable and are expected to increase significantly in times of heavy rainfall and flooding.

#### *Production and disposal of cadmium-containing materials*

The estimated cadmium discharge to waters from the manufacture of cadmium-containing articles,  $108 \text{ t y}^{-1}$ , was about four times higher than the values given by both Rauhut (1978) and Van Wambeke (1979). Electroplating facilities accounted for the largest input of cadmium to aquatic systems at  $61 \text{ t y}^{-1}$ . This quantity was greater than the combined total from the three other major users of cadmium. Pigment and stabilizer losses to water together totalled  $33 \text{ t y}^{-1}$ , compared with the much lower estimate of  $4 \text{ t y}^{-1}$  provided by Rauhut. Similarly, the present study's estimate of aquatic discharges from the nickel-cadmium battery industry amounted to about  $14 \text{ t Cd y}^{-1}$  compared with Rauhut's value of  $4 \text{ t y}^{-1}$ . Further investigations at a practical level are required in order to verify

these inputs of cadmium to water. This is considered to be an important need, because unlike the other major sources of aquatic cadmium (zinc production, non-ferrous metal mines, and phosphate fertilizer wastes) a large proportion of the cadmium discharged from this sector is expected to enter sewage treatment systems.

The disposal of wastes arising from the manufacture of cadmium-containing articles results in an estimated discharge of about  $90 \text{ t Cd y}^{-1}$  to landfill. Rauhut's estimate for this value was somewhat lower at  $51 \text{ t y}^{-1}$ . Nevertheless, both studies indicate that wastes from electroplating facilities make the largest contribution to this discharge; the value assigned by the present study is  $60 \text{ t y}^{-1}$  compared with a total of  $29 \text{ t y}^{-1}$  from stabilizer and nickel-cadmium battery manufacture. The  $90 \text{ t}$  cadmium associated with the manufacture of cadmium-containing articles will presumably be disposed of to controlled landfills. Nevertheless, the markedly elevated concentrations of these wastes suggests a high potential for leaching to adjacent watercourses. The regular monitoring of such sites will reveal whether this is occurring. In contrast, the  $653 \text{ t}$  cadmium assumed to be disposed of to landfill with discarded electroplated items will, in the short term at least, show a low rate of leaching from disposal sites.

The recycling of cadmium-plated steel scrap is largely responsible for the emissions of cadmium from the steel industry and was estimated to result in a large input ( $434 \text{ t y}^{-1}$ ) of cadmium into this sector. Neither Rauhut or Van Wambeke provide any estimates for the quantity of cadmium entering the steel industry via scrap recycling.

This study has assumed that all of the plastic wastes which contain cadmium pigments and stabilizers will enter the waste disposal pathway. The absence of any recycling of cadmium in these products is a situation which is unlikely to change in the future.

### *The iron and steel industries*

This study found that the production of iron and steel is the single largest source of atmospheric cadmium and accounts for 30 per cent of the total emissions from human activities. Iron production was assumed to result in an atmospheric emission of about  $11 \text{ t y}^{-1}$  in the EC. Rauhut's (1978) estimate was much higher at  $89 \text{ t y}^{-1}$ . The latter value is considered to be too high and is not supported by the findings of the study described in Appendix 2. Moreover, in the case of steel production, Rauhut considered only open-hearth steelmaking, a process declining

in importance in the EC. Thus, no estimate of atmospheric emissions from electric arc or basic oxygen furnaces was made by this earlier estimate. Van Wambeke assigned a total of 130–200 t cadmium emitted to the atmosphere from steel production in 1975; this compares with the present study's estimate of 24 t.

The quantity of cadmium discharged to landfills from iron and steel production,  $350 \text{ t y}^{-1}$ , is greater than the contribution from zinc production. For reasons which are unclear, Rauhut (1978) did not assign any quantity of cadmium to solid wastes from this source. Van Wambeke (1979) estimated that 330–400 t cadmium would be discharged in solid wastes from steel production alone, with an additional 200–300 t  $\text{Cd y}^{-1}$  derived from iron production.

Solid wastes collected from electric arc furnaces are invariably disposed of to controlled landfills. Nevertheless, the high cadmium concentrations associated with these wastes indicates that leaching may occur at these sites. Rauhut (1978) suggested that the cadmium collected in dusts from steelworks can be recycled in zinc–cadmium refineries. This, however, is not the case and there is negligible recycling of arrested dusts from steel furnaces in the EC.

#### *Fuel combustion*

The atmospheric emission estimate for hard coal combustion totalled less than  $6 \text{ t y}^{-1}$ . This figure is much lower than the values assigned by previous EC studies: Rauhut (1978) provided a figure of  $70 \text{ t y}^{-1}$ , Van Enk (1979)  $450 \text{ t y}^{-1}$ , and Van Wambeke (1979)  $60 \text{ t y}^{-1}$ . All these earlier estimates are considered to be too large. The present study's estimate was computed after taking into consideration the results of in-stack measurements of cadmium's partitioning in coal-fired power plants. The emission factor used was similar to that employed by Yost (1979) and Nriagu (1979).

The quantity of cadmium associated with coal ashes was estimated to be 390 t. For the purposes of this study, all this cadmium was assumed to enter landfill sites. This is an oversimplification as a significant proportion will be utilized on a commercial basis as construction material. Of the previous EC studies, Rauhut did not consider cadmium in solid wastes from coal combustion while Van Wambeke's estimate was similar to that of the present study, at  $340 \text{ t Cd y}^{-1}$ .

Atmospheric emissions of cadmium from oil combustion in the EC have previously been estimated at  $5 \text{ t y}^{-1}$  by Van Wambeke and  $21 \text{ t y}^{-1}$

by Rauhut. Rauhut's estimate was based on an average cadmium level in oil which was 10 times higher than assumed by this study. Additionally, Rauhut assumed that emissions from facilities burning oil would be uncontrolled; this is generally not the case.

In summary, it would appear that the rôle of fuel combustion as a source of atmospheric cadmium in the EC has been exaggerated in the past. The present study's total atmospheric emission of cadmium from fuel combustion amounts to  $8.5 \text{ t y}^{-1}$ , compared with Rauhut's estimate of  $106 \text{ t y}^{-1}$ , Van Wambeke's of  $80 \text{ t y}^{-1}$  and Van Enk's of  $470 \text{ t y}^{-1}$ .

### *Waste disposal*

The incineration of refuse accounts for the second largest emission of cadmium to the atmosphere of the EC ( $31 \text{ t y}^{-1}$ ) and the largest input of cadmium to landfill sites ( $1,434 \text{ t y}^{-1}$ ). Previous EC studies have assigned a roughly similar value to the quantity of cadmium released to the atmosphere from this source; Van Wambeke gave an estimate of  $30 \text{ t y}^{-1}$  and Rauhut  $18 \text{ t y}^{-1}$ .

### *Sewage sludge*

The quantity of cadmium contained in sewage sludge from the EC has been estimated to be  $165 \text{ t y}^{-1}$ . This value is higher than the figure of  $70\text{--}100 \text{ t Cd y}^{-1}$  given by Van Wambeke. Rauhut did not consider sewage sludge in his inventory of cadmium emissions in the EC.

### *Phosphate fertilizers*

Disposal of gypsum waste from phosphate fertilizer manufacture results in a significant discharge of cadmium to aquatic systems. In most cases, however, these discharges take place in coastal areas and do not enter sewage treatment systems. About  $300 \text{ t Cd y}^{-1}$  were estimated to be applied to land as a consequence of fertilizer use in the EC. This value is similar to Van Wambeke's estimate of  $270 \text{ t}$  cadmium present in fertilizer products and residues. However, it is not clear from the latter estimate whether all of the  $270 \text{ t Cd y}^{-1}$  is assumed to be applied to agricultural land.

#### *1.11.5 Future trends*

Table 18 summarizes the predicted discharges of cadmium to the EC environment for 1990 and 2000.

**Table 18 Summary of predicted cadmium inputs to the EC environment in 1990/2000 ( $\text{t y}^{-1}$ )**

| Source                                     | Compartment |             |         |
|--------------------------------------------|-------------|-------------|---------|
|                                            | Air         | Land        | Water   |
| Volcanic action                            | 20/20       | ND          | ND      |
| Non-ferrous metal production               |             |             |         |
| Zinc and cadmium                           | 25/28       | 240/286     | 60/70   |
| Copper                                     | 7/8         | 20/25       |         |
| Lead                                       | 8/9         | 45/49       | 22/25   |
| Production of cadmium-containing materials | 3/3         | 104/116     | 117/124 |
| Iron and steel production                  | 38/46       | 479/583     | ND      |
| Fuel combustion                            |             |             |         |
| Coal and lignite                           | 9/14        | 446/667     | ND      |
| Oil and gas                                | <1/<1       | 18/18       | —       |
| Waste disposal                             | 31/31       | 1,434/1,434 | ND      |
| Sewage sludge disposal                     | 5/9         | 161/195     | 29/36   |
| Phosphate fertilizers                      | —           | 346/346     | 62/62   |
| Totals                                     | 146/169     | 3,293/3,719 | 290/317 |

ND—Not determined.

Significant increases in atmospheric inputs of cadmium are forecast for zinc production, iron and steel production, and sludge incineration. Greater reliance upon coal as a source of energy in the future has been predicted to result in a near doubling of atmospheric cadmium emissions from this sector by the year 2000. The iron and steel industries, together with coal combustion, are the major sources responsible for the large increases in cadmium discharged to land by 2000. The data for inputs to water are incomplete but zinc production is forecast to increase discharges to this medium in the future.

## 2 Environmental concentrations of cadmium in the EC

### 2.1 Introduction

The environmental distribution of cadmium has been the subject of numerous investigations in the EC. Several studies have attempted to summarize the information generated, at both the national level (DOE

Table 19 Representative atmospheric cadmium concentrations from selected localities in the EC

| Area and locality          | Concentration<br>(ng m <sup>-3</sup> ) | Comment                                          | Reference                        |
|----------------------------|----------------------------------------|--------------------------------------------------|----------------------------------|
| <b>Rural</b>               |                                        |                                                  |                                  |
| U.K.                       | 1-2.7                                  | Range of annual averages from 7 sites            | Cawse (1977)                     |
| Botrange, Belgium          | 4-6                                    | Range of annual averages, 1972-77                | Kretzschmar <i>et al.</i> (1980) |
| Witteveen, The Netherlands | 5                                      | —                                                | Minderhoud and Boogerd (1975)    |
| Corviglia, F.R.G.          | 2.1                                    | —                                                | Muller and Beilke (1977)         |
| <b>Urban</b>               |                                        |                                                  |                                  |
| London, U.K.               | 3.6-8.9                                | Range of annual averages from 5 sites in Lambeth | Branson and Pattenden (1979)     |
| Brussels, Belgium          | 8-15                                   | Range of annual averages, 1972-77                | Kretzschmar <i>et al.</i> (1980) |
| Paris, France              | 20                                     | —                                                | Belot <i>et al.</i> (1971)       |
| Frankfurt, F.R.G.          | 6.1                                    | —                                                | Muller and Beilke (1977)         |
| <b>Industrial</b>          |                                        |                                                  |                                  |
| Liège, Belgium             | 18-54                                  | Range of annual averages, 1972-77                | Kretzschmar <i>et al.</i> (1980) |
| London, U.K.               | 200-11,000                             | ≈15 m from lead recovery plant                   | Muskett <i>et al.</i> (1979)     |
| Walsall, U.K.              | 16.5-40                                | ½ km from copper refinery                        | Playford and Pattenden (1979)    |

1980; Miljøministeriet 1980) and for the European Community as a whole (CEC 1978). For this reason, there is no intention here to provide an exhaustive review of all available environmental monitoring data from the EC. Instead, this section considers the cadmium concentrations in those environmental media associated with human exposure and the levels and significance of cadmium in terrestrial organisms.

## 2.2 Atmospheric concentrations

Table 19 shows atmospheric cadmium concentrations measured at various sites in the European Community. Airborne cadmium levels in rural localities are very low and their measurement can present analytical difficulties, particularly for neutron activation techniques. Cadmium concentrations in urban areas are higher than in rural localities but the difference is not marked and in general corresponds to an increase of less than one order of magnitude. Air concentrations in industrial areas or at sites adjacent to point sources can be much greater, as illustrated by the extreme value of  $11 \mu\text{g m}^{-3}$ , measured near a small lead refinery in London, U.K. (Muskett, Roberts and Page 1979). Elevated values may also be encountered in close vicinity to volcanoes; Buat-Ménard and Arnold (1978) recorded a level of  $30 \mu\text{g m}^{-3}$  above a hot vent of Mount Etna.

It has generally been assumed in the past that airborne cadmium concentrations, like those provided in Table 19, are valid measures of respiratory exposure in humans. However, air sampling often occurs on roof-tops and at other sites located more than 1–2 m above ground, the height at which adults normally breath. The resuspension of settled dusts, particularly in urban areas, may produce airborne cadmium concentrations at 1–2 m which are significantly greater than those measured at roof-top height. There is thus a possibility that human respiratory exposure is underestimated in this way. This, however, is only considered to be of importance in those urban locations where dust cadmium levels are elevated, like those sites adjacent to point sources of atmospheric cadmium (see 2.4).

## 2.3 Deposition of cadmium from the atmosphere

The removal of airborne cadmium occurs by dry deposition and by precipitation. Total (dry plus wet) deposition rates for cadmium from various localities in the EC are depicted in Table 20. Classification of

**Table 20 Total deposition of cadmium at selected localities in the EC**

| Area and locality      | Deposition rate<br>(g ha <sup>-1</sup> y <sup>-1</sup> ) | Comment                          | Reference                     |
|------------------------|----------------------------------------------------------|----------------------------------|-------------------------------|
| <b>Rural</b>           |                                                          |                                  |                               |
| Norfolk, U.K.          | 2.6                                                      | —                                | Horler and Barbour (1979)     |
| Texel, The Netherlands | 2.9                                                      | —                                | P. W. van N. H.* (1975)       |
| Belgium                | 19                                                       | Average of 19 agricultural sites | Navarre <i>et al.</i> (1980)  |
| Denmark                | 3.0                                                      | Assumed background value         | Miljøministeriet (1980)       |
| <b>Urban</b>           |                                                          |                                  |                               |
| London, U.K.           | 24.7–29.6                                                | Range from three sites           | Harrison <i>et al.</i> (1975) |
| Göttingen, F.R.G.      | 3.9                                                      | —                                | Ruppert (1975)                |
| <b>Industrial</b>      |                                                          |                                  |                               |
| Walsall, U.K.          | 150                                                      | < 1 km from Cu refinery          | Cawse (1979)                  |
| Liège, Belgium         | 82                                                       | —                                | Navarre <i>et al.</i> (1980)  |
| Nordenham, F.R.G.      | 135                                                      | Close to zinc smelter            | Quoted in CEC (1978, page 31) |

\* Provinciale Waterstaat van Noord Holland.

these localities reveals that in most cases, deposition rates, like air concentrations, increase in the order rural < urban < industrial. The deposition rates for most rural locations given in Table 20 indicate that 3 g ha<sup>-1</sup> y<sup>-1</sup> is a representative value for atmospheric inputs to agricultural soils in rural areas of the EC. The average deposition rate of 19 g ha<sup>-1</sup> y<sup>-1</sup> reported for rural agricultural areas in Belgium (Navarre, Ronneau and Priest 1980) is markedly higher than other Member States in the EC. This disparity may correspond to the importance of the non-ferrous metal industry in Belgium, but further work is required in order to investigate these anomalously high deposition rates.

There is only limited information available on cadmium deposition rates in those urban areas in the EC which lack any cadmium-using

industries. Data from the two sites shown in Table 20 indicate that a large range of values may be encountered in such locations; at the U.K. site, urban values are an order of magnitude higher than rural areas.

Deposition rates of cadmium can be very high near to point sources of the metal, as illustrated by the two values from the U.K. and F.R.G. shown in Table 20. In general, there is a rapid decline in deposition rate with distance from the cadmium source. However, in an urban area like Liège, the high density of non-ferrous metal works results in a city-wide elevation of cadmium deposition rates (Roels, Lauwerys, Buchet and Bernard 1981).

#### 2.4 Cadmium in dusts and soils

Little attention has been paid in the EC to the presence of cadmium in deposited dusts from urban areas. This contrasts with the situation for lead, where numerous studies have considered that the ingestion of urban dusts may be an important source of exposure in children (e.g. see Duggan 1980).

One of the few published studies originating from the EC reported an average cadmium level in U.K. street dusts from an urban locality of  $4.6 \mu\text{g g}^{-1}$  (Harrison 1979). Household dusts from the same location contained over  $10 \mu\text{g g}^{-1}$ , more than double that found for external dusts. The bioavailability of cadmium to humans from these dusts is expected to be high, as over 60 per cent of the metal was extracted with 0.07 M hydrochloric acid.

The cadmium present in external dusts is derived in part from the abrasion of motor vehicle tyres (Lagerwerff and Specht 1970), but refuse incineration may also make a significant contribution (Greenberg *et al.* 1978). The reason for the high cadmium levels in U.K. household dusts is not clear but this finding may be associated with the abrasion of the rubber backing on carpets; one study reported that the rubber used for this purpose in the U.S.A. contained  $3,000 \mu\text{g g}^{-1}$  cadmium (Solomon and Hartford 1976). It is not known at present whether the rubber used for backing carpets in any Member States of the EC also contains such high cadmium levels.

Elevated cadmium concentrations, averaging  $193 \mu\text{g g}^{-1}$ , have been measured in household dusts from a site adjacent to a lead refinery (Muskett, Roberts and Page 1979). The ingestion of such dusts may result in a significant increase in cadmium exposure. Additionally, the

resuspension of these dusts may also result in enhanced respiratory exposure at such localities.

The background concentrations of cadmium in soils will vary to some extent according to the parent rock from which they are derived. Cadmium concentrations in the majority of agricultural soils from the EC probably lie within the range of  $0.1\text{--}2.0\ \mu\text{g g}^{-1}$ , with most values being less than  $1.0\ \mu\text{g g}^{-1}$  (Archer 1980; Davis and Coker 1980; Miljøministeriet 1980).

Elevated concentrations of cadmium in soils can originate from natural sources as well as from human activities. Soils derived from Carboniferous black shales, for example, contain naturally elevated cadmium levels.

Past mining activities in certain mineralized areas of the EC have led to significant cadmium contamination of the surrounding soils. The cadmium concentrations encountered in these soils are variable, being dependent upon the cadmium status of the ore body in question. The unusually high soil cadmium levels at one site in the U.K., Shipham, prompted investigations into the exposure levels and health of the local community. The results of these studies are summarized in Appendix 6. Soil cadmium concentrations similar to those found in Shipham gardens have also been recorded at sites in Wales (Johnson, Roberts, Hutton and Inskip 1978), Belgium and the F.R.G. (Simon 1977).

Atmospheric emissions of cadmium from non-ferrous metal smelters and refineries produce elevated cadmium levels in local soils but these decline rapidly with distance from the source. The concentrations encountered are dependent upon the size of atmospheric cadmium emissions, both past and present, as well as factors such as prevailing wind direction, stack height and local topography. Smelter wastes may be an additional source of soil contamination at those localities with a long history of non-ferrous metal smelting, as such wastes were in the past often dumped in or near to the works themselves. Examples of sites in the EC where contamination from such wastes is a persistent problem are the Swansea Valley, Wales (Welsh Office 1975) and Prayon, Belgium (Simon 1977).

The possible contamination of soils from other point sources of airborne cadmium has not been investigated fully in the EC. In the case of coal combustion, several American studies have measured soil cadmium concentrations in the vicinity of large coal-fired power plants. Klein and Russel (1973) claimed that soils around a Michigan plant

were enriched with cadmium but the data were not subjected to statistical analysis. In contrast, an extensive survey of the Allen Steam Plant in Memphis detected no increases in the cadmium levels of surrounding soils (Bolton *et al.* 1973; Lyon 1977). In view of the results of the latter study, it is considered unlikely that significant increases in cadmium levels will be detectable in soils surrounding EC power plants.

Apparently, there are no published studies of soil cadmium levels in the vicinity of refuse incinerators in the EC. These facilities emit larger quantities of airborne cadmium than coal power plants and it is thus conceivable that cadmium levels in surrounding soils will be elevated above background.

A source of soil contamination which may be of potential significance in certain industrial regions of the EC arises from the practice of dumping dredged river sediments onto adjacent agricultural land. One study in the F.R.G. (Mack and Schmid 1979) revealed that sediments from the River Neckar contain elevated cadmium levels; this contamination was thought to originate from a local cadmium pigment plant. Agricultural land which had received applications of these soil sediments contained  $65 \mu\text{g g}^{-1}$  cadmium.

The application of phosphate fertilizers results in a significant input of cadmium to agricultural soils; total discharges from this source were estimated earlier for individual Member States and shown in Table 15. In order to appreciate fully the significance of this source, the values given in Table 15 require conversion to annual rates of cadmium input to arable land. These input rates, shown in Table 21, vary considerably, reflecting the differences in fertilizer application rates between Member States as well as differences in the cadmium content of the fertilizers used. The highest cadmium input rate from fertilizer use in the EC occurs in Belgium, mainly as a consequence of the high application rates employed in this country (OECD 1973). This finding is particularly noteworthy as atmospheric deposition rates to agricultural land in Belgium are apparently also the highest in the region (Table 20).

Other estimates of cadmium inputs to agricultural land from phosphate fertilizers have been made for Denmark (Miljøministeriet 1980) and the F.R.G. (Jung, Isermann and Henjes 1979). The Danish figure of  $3 \text{ g Cd ha}^{-1} \text{ y}^{-1}$  is similar to that estimated in Table 21. In contrast, the figure of  $1.67 \text{ g ha}^{-1} \text{ y}^{-1}$  given by Jung *et al.* (1979) for the F.R.G. is significantly lower than the value of  $4.6 \text{ g ha}^{-1} \text{ y}^{-1}$  estimated in the present study. This disparity arises from the use of lower application

**Table 21** Estimated phosphate fertilizer use and corresponding annual cadmium inputs to arable land in Member States of the EC

| Country             | Annual fertilizer input <sup>a</sup><br>(kg P <sub>2</sub> O <sub>5</sub> ha <sup>-1</sup><br>arable land) | "Average" Cd content <sup>b</sup><br>(g t <sup>-1</sup> P <sub>2</sub> O <sub>5</sub> ) | Annual Cd input to<br>arable land<br>(g ha <sup>-1</sup> ) |
|---------------------|------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------|------------------------------------------------------------|
| Italy               | 29.8                                                                                                       | 55                                                                                      | 1.6                                                        |
| The Netherlands     | 57                                                                                                         | 80                                                                                      | 4.6                                                        |
| U.K.                | 64                                                                                                         | 101                                                                                     | 6.5                                                        |
| F.R.G.              | 96                                                                                                         | 48                                                                                      | 4.6                                                        |
| Belgium             | 144                                                                                                        | 65                                                                                      | 9.4                                                        |
| France              | 76                                                                                                         | 71                                                                                      | 5.4                                                        |
| Denmark             | 46                                                                                                         | 52                                                                                      | 2.4                                                        |
| Republic of Ireland | 124                                                                                                        | 53                                                                                      | 6.6                                                        |
| EC average          |                                                                                                            |                                                                                         | 5.1 ± 2.5                                                  |

<sup>a</sup> 1967/68 data, taken from OECD (1973). The figure for Italy may be unrealistic as fertilizer use in 1979/80 has doubled since 1967/68.

<sup>b</sup> Source: Appendix 5. "Average" Cd content calculated from 1979/80 fertilizer consumption and total Cd discharge from this source.

rates and assumed cadmium levels of phosphate fertilizer by Jung *et al.* compared with the present study. A document recently published by the Umweltbundesamt (1980) has assigned a cadmium input rate of 5–6 g ha<sup>-1</sup> y<sup>-1</sup> to agricultural land from phosphate fertilizer use in the F.R.G. This figure is much closer to the value given in Table 21 than that estimated by Jung *et al.* (1979).

Table 21 reveals that the quantity of cadmium entering agricultural soils from phosphate fertilizer use is similar to, if not greater than, the total input from all atmospheric sources of cadmium. Despite this relatively large contribution, the annual input of cadmium from fertilizers is some 100 times lower than the cadmium burden already present in soils. It is therefore not surprising that difficulties exist in detecting the influence of fertilizer applications upon cadmium levels in agricultural soils. Nevertheless, soils from two localities known to have received long-term applications of phosphate fertilizers were reported to contain elevated cadmium levels compared with controls (Williams and David 1973; Mulla, Page and Ganje 1980). However, the actual soil levels encountered at both contaminated sites were by no means high and fall within the range of values given earlier for normal agricultural soils.

The addition of sewage sludge to agricultural land often occurs at higher rates of application than those employed for phosphate fertilizers. Not surprisingly, this can result in much larger increases in soil cadmium levels. Indeed, certain agricultural areas receiving sewage from industrialized cities in both the EC and U.S.A. have been reported to contain markedly elevated soil cadmium levels (Davis and Coker 1980; Williams, Vlamis, Pukite and Corey 1980). On a national or regional level however, sewage sludge additions represent a much smaller input of cadmium to agricultural soils than do either phosphate fertilizers or the deposition of airborne cadmium. One estimate from Denmark (Hovmand 1980) indicates that the application of sewage sludge accounts for only 2 per cent of the total input of cadmium to agricultural land.

The significance of cadmium inputs to soils from the sources discussed above has in the past been considered only in terms of the increase in the burden of total soil cadmium. This approach assumes that cadmium inputs from different sources display an equal potential for biological uptake. There is, however, evidence that cadmium inputs from phosphate fertilizers are initially less available for plant uptake than inputs from sewage sludge (Webber 1979; Mulla *et al.* 1980). Additionally, soil cadmium which originated from atmospheric deposition is expected to show a greater potential for plant uptake than cadmium derived from mining activities, where a proportion of the metal will be in the form of unweathered mineral matter.

A further factor which may be of importance when assessing the long-term significance of cadmium inputs to agricultural land is the possibility that phosphate additions may actually reduce the bioavailability of soil cadmium. There is certainly experimental evidence that soil zinc availability to plants is reduced by phosphate applications (Badanur and Venkata Rao 1972; Berrow and Burridge 1979). Furthermore, the cadmium content of plants grown on some soils with a long history of phosphate applications and elevated cadmium levels were actually lower than in plants grown on similar soils with no fertilizer additions (Williams and David 1976; Mulla *et al.* 1980).

## 2.5 Cadmium uptake by terrestrial organisms

### 2.5.1 Cadmium in plants

Several reports describing the factors which influence cadmium uptake by plants have recently become available (Cataldo and Wildung 1978;

Davis and Coker 1980; Peterson and Alloway 1980). For this reason, it is not intended to present a detailed discussion of those parameters which determine plant uptake of cadmium. However, attention will be paid to those factors employed in Section 4 for the modelling of future trends in soil-plant cadmium relations.

Cadmium in soils is distributed between a number of pools of which only the cadmium in soil solution is thought to be available for plant uptake directly. The distribution of cadmium between the soil and soil solution is described by the adsorption coefficient ( $\alpha$ ), defined as the ratio of the concentration of cadmium in soil solution and the concentration in the soil. A soil with a large adsorption coefficient will display a relatively high potential for plant uptake of cadmium. Factors which determine the soil solution concentration will thus influence plant uptake of the metal. Such factors include soil composition, soil cadmium content and soil pH. Soil pH in particular has a pronounced influence upon the availability of soil cadmium to plants. The lower the pH of the soil, the more cadmium is released to soil solution, resulting in a greater availability for plant uptake.

Cadmium levels in a range of edible plant species from uncontaminated localities are shown in Table 22. Concentrations vary widely between species but most values fall within the range of 0.05–0.2  $\mu\text{g g}^{-1}$ . Exceptions include leafy vegetables such as lettuce which contain significantly higher levels, while values in grain crops are lower. Pronounced differences in cadmium uptake can also exist between different genotypes of the same species (John and Van Laerhoven 1976). Additionally, plant

**Table 22 Cadmium concentrations in food plants from uncontaminated soils**

| Species      | Cadmium concentration<br>( $\mu\text{g g}^{-1}$ wet weight) |           | Authority                       |
|--------------|-------------------------------------------------------------|-----------|---------------------------------|
|              | Mean                                                        | Range     |                                 |
| Wheat grains | 0.068                                                       | —         | Kjellström <i>et al.</i> (1975) |
| Cabbage      | 0.04                                                        | 0.01–0.15 | Thomas <i>et al.</i> (1972)     |
| Potato       | 0.08                                                        | 0.01–0.17 | Thomas <i>et al.</i> (1972)     |
| Carrot       | 0.13                                                        | 0.09–0.22 | Thomas <i>et al.</i> (1972)     |
| Tomato       | 0.02                                                        | 0.01–0.08 | Thomas <i>et al.</i> (1972)     |
| Lettuce      | —                                                           | 0.01–1.81 | Lindsay (1980)                  |
| Leeks        | —                                                           | 0.02–0.23 | Lindsay (1980)                  |

cadmium concentrations often vary according to season, with higher values being reported in autumn and winter (Davis and Coker 1980).

The selectivity coefficient ( $S$ ) is used to describe cadmium uptake by plants and is defined as the ratio of actual cadmium uptake and the expected uptake, calculated from the water taken up and the soil solution concentration of cadmium. Experimental difficulties exist in measuring the selectivity coefficient but considerable differences have been noted between different species (Poelstra, Frissel and El-Bassam, 1979).

The cadmium content of different parts of a particular plant species often vary significantly. In general, the concentration decreases in the order roots > stem > leaves > seeds or fruiting body (Jarvis, Jones and Hopper 1976). Distributional differences of cadmium between plant parts will be reflected by differences in the selectivity coefficient. In soya bean, for example, Cunningham, Collins and Hutchinson (1975) found a selectivity coefficient of 8.6 in roots, 4.5 in the stem, and 1.4 in the leaves.

Plants growing in cadmium contaminated localities can accumulate elevated levels of the metal. Table 23 depicts values in selected species from such sites. Much of the data in Table 23 refer to levels in lettuce, a species which accumulates more cadmium than many other plants. This point is illustrated by the findings of a survey at Shipham in the U.K., where cadmium levels in lettuce were elevated, while those in French beans and tomatoes were similar to background values. Similarly, lettuce grown in sewage sludge amended soil contained enhanced cadmium concentrations but corn grain showed no significant difference in cadmium content compared with control values.

Caution should be exercised when drawing comparisons between the cadmium levels reported in lettuce from the contaminated sites in Table 23. This is because the lettuce varieties used by these different studies were not specified and up to a seventyfold difference exists in cadmium uptake between different cultivars of this species (John and Van Laerhoven 1976). Nevertheless, it is of interest to note that cadmium levels in lettuce grown near to a copper refinery were similar to those at Shipham, despite the considerably lower soil cadmium content at the former site. This difference may be the consequence of a greater availability of soil cadmium to plants from atmospheric inputs, but surface contamination of leaves by airborne cadmium may also contribute to the cadmium burden of lettuce at the smelter site.

Table 23 Cadmium concentrations in selected plant species from contaminated sites

| Species                     | Cadmium level ( $\mu\text{g g}^{-1}$ ) |                  |                         | Cadmium source               | Authority |
|-----------------------------|----------------------------------------|------------------|-------------------------|------------------------------|-----------|
|                             | Plant<br>(wet wt)                      | Soil<br>(dry wt) |                         |                              |           |
| Lettuce                     | 0.38-0.75 <sup>a</sup>                 | <4-9.2           | Copper refinery         | Cawse (1979)                 |           |
| Lettuce                     | 0.69                                   | 87 (2-520)       | Zinc mine               | Shiphams Survey Ctte (1980)  |           |
| Tomatoes                    | 0.06                                   | 87 (2-520)       | Zinc mine               | Shiphams Survey Ctte (1980)  |           |
| French beans                | 0.02                                   | 87 (2-520)       | Zinc mine               | Shiphams Survey Ctte (1980)  |           |
| Lettuce                     | 0.35                                   | 65               | Contaminated sediments  | Mack and Schmid (1979)       |           |
| Lettuce                     | 0.003                                  | —                | Control                 | Mack and Schmid (1979)       |           |
| Lettuce                     | 0.50                                   | 5-10             | Sewage sludge           | Lindsay (1979)               |           |
|                             | (0.24-0.94)                            |                  |                         |                              |           |
| Corn grain <sup>b</sup>     | 0.06                                   | 5.2              | Sewage sludge           | Webber and Beauchamp (1979)  |           |
| Corn grain                  | 0.03                                   | 0.48             | Control                 | Webber and Beauchamp (1979)  |           |
| Rye-grass <i>Lolium</i>     | 50.0                                   | —                | 0.3 km from Zn smelter  | Burkitt <i>et al.</i> (1972) |           |
| <i>perenne</i> <sup>b</sup> | 10.8                                   | —                | 1.9 km from Zn smelter  | Burkitt <i>et al.</i> (1972) |           |
|                             | 1.8                                    | —                | 11.3 km from Zn smelter | Burkitt <i>et al.</i> (1972) |           |

<sup>a</sup> Converted from dry wt by  $\times 0.1$ .

<sup>b</sup> Plant cadmium concentration in  $\mu\text{g g}^{-1}$  dry wt.

The decline in cadmium contamination with distance from a point source is illustrated by the decreasing cadmium content of rye-grass samples collected at increasing distances from a zinc smelter in the U.K.

### 2.5.2 *Effects of environmental cadmium on plants*

Toxic effects of cadmium on plants, such as stunting, chlorosis and necrosis, can be demonstrated under experimental conditions, but their occurrence in environmentally exposed vegetation is unusual. In general, it is rare to observe cadmium-induced toxic effects in vegetation from contaminated areas simply because cadmium is rarely found in soils at levels which are overtly toxic to plants.

One effect of possible ecological importance—the development of cadmium-tolerant ecotypes in cadmium-contaminated areas—has only recently been recorded. The first report on this subject concerned populations of two grass species from a zinc mine and near to a zinc smelter in Belgium (Simon 1977). There is also some evidence that soils near to a zinc smelter in the U.K. contain greater proportions of cadmium-tolerant microfungi populations compared with uncontaminated soils (Martin, Coughtrey, Shales and Little 1980). Similarly, phylloplane populations growing on contaminated plants from around the same smelter displayed greater cadmium tolerance than those from control plants (Gingell, Campbell and Martin 1976). Additionally, the leaf surface microflora from these contaminated plants was reduced in both abundance and species diversity (Gingell *et al.* 1976).

### 2.5.3 *Cadmium in animals*

Terrestrial invertebrates are important components of food chains and are generally considered to play a key role in the cycling of minerals. Nevertheless, the accumulation, tissue distribution and possible effects of cadmium in this group have only recently received attention.

The tissue distribution of cadmium in terrestrial invertebrates has been examined in a limited number of species; Table 24(a) depicts values obtained in the woodlouse *Oniscus asellus* and the snail *Cepaea hortensis* from contaminated sites in the U.K. Highest levels were found in the hepatopancreas of the isopod and the digestive gland of the gastropod. Relatively low levels were present in the hard parts (exoskeleton) of *Oniscus*. A similar accumulation of metals in the digestive gland of marine molluscs has been reported on a number of occasions (Bryan 1973; Ireland and Wootton 1977).

**Table 24** Cadmium concentrations in various invertebrates from contaminated localities in the U.K. ( $\mu\text{g g}^{-1}$  dry wt)

| <b>(a) Tissue distribution in an isopod and a gastropod</b>       |              |               |                 |        |
|-------------------------------------------------------------------|--------------|---------------|-----------------|--------|
|                                                                   | Whole animal | Hard parts    | Hepatopancreas  |        |
| Woodlouse                                                         |              |               |                 |        |
| <i>Oniscus asellus</i>                                            | 87.7         | 20.1          | 786.7           |        |
|                                                                   | Whole animal | Head and foot | Digestive gland | Kidney |
| Snail*                                                            |              |               |                 |        |
| <i>Cepaea hortensis</i>                                           | 14.1         | 3.6           | 40.3            | 12.8   |
| <b>(b) Total body concentrations in three invertebrate groups</b> |              |               |                 |        |
|                                                                   | Whole animal |               |                 |        |
| Slugs                                                             |              |               |                 |        |
| <i>Arion fasciatus</i>                                            | 28.7         |               |                 |        |
| <i>Arion hortensis</i>                                            | 56.5         |               |                 |        |
| <i>Agriolimax reticulatus</i>                                     | 43.0         |               |                 |        |
| Snails                                                            |              |               |                 |        |
| <i>Helix aspersa</i>                                              | 62.4         |               |                 |        |
| <i>Clausilia bidentata</i>                                        | 76.1         |               |                 |        |
| Earthworms (species unknown)                                      | 144          |               |                 |        |

Source: Martin *et al.* (1980).

\* Williamson (1980).

Table 24(b) shows the body burdens of cadmium found in various invertebrate species from a contaminated woodland site in the U.K. The marked accumulation of cadmium in earthworms is thought to result from the ingestion of large quantities of soil and leaf litter. Elevated cadmium levels have also been reported in earthworms from roadside soils (Gish and Christensen 1973), urban soils and from sewage sludge amended soils (Andersen 1979). Cadmium concentrations in earthworms from an urban garden in Denmark averaged  $25 \mu\text{g g}^{-1}$ , compared with soil cadmium levels of  $0.29\text{--}1.0 \mu\text{g g}^{-1}$ , representing concentration factors of 20–100 (Andersen 1979).

Cadmium concentration factors ( $\mu\text{g g}^{-1}$  animal:  $\mu\text{g g}^{-1}$  presumed diet) have also been computed for the invertebrates shown in Table 24(b) (Martin *et al.* 1980). The ratios obtained ranged from 4.6–42 and in

every case were larger than those for lead and zinc, indicating that cadmium has a greater potential for accumulation through food chains than the other two metals. Some evidence for this suggestion has been provided by a study of metal levels in invertebrates and their estimated diets from an abandoned lead-zinc mine in the U.K. (Roberts and Johnson 1978). Cadmium concentrations in herbivorous invertebrates were somewhat higher than in vegetation but, more important, cadmium levels in carnivorous invertebrates were markedly higher than in the herbivores. No such accumulation was observed for either lead or zinc. The preferential accumulation of cadmium in the soft parts of invertebrates (Table 24(a)) may account for the apparent mobility of cadmium in this food chain.

These studies suggest that cadmium can accumulate through invertebrate food chains, at least in grossly contaminated localities. However, the technique employed to demonstrate this effect (a simple cadmium concentration ratio) takes no account of the factors which may also influence the cadmium burden of a particular invertebrate. In particular, invertebrates display an age-dependent accumulation of cadmium (Williamson 1980) and thus the age of an invertebrate sampled will influence its cadmium content. Additionally, different invertebrate groups exhibit variable calcium requirements and this may also affect the uptake of cadmium from the gut, as the absorption of the two metals appears to be linked.

Generally, cadmium burdens in terrestrial vertebrates from uncontaminated localities are low. Similarly, most domesticated species reared for human consumption are slaughtered at an early age, preventing any significant accumulation of cadmium. Horses are an exception to this practice and older individuals can contain markedly elevated renal cadmium concentrations (Friberg, Piscator, Nordberg and Kjellström 1974).

Mammals and birds exhibit similar tissue distributions of cadmium after environmental exposure, with highest concentrations in kidney, substantial quantities in liver and much lower levels in all other tissues (Johnson *et al.* 1978; Hutton and Goodman 1980).

Only recently has any attention been paid to the accumulation of cadmium by vertebrates living in contaminated localities. Small mammal species from both an abandoned lead-zinc mine and from a zinc smelter complex in the U.K. contained elevated cadmium burdens (Roberts and Johnson 1978; Johnson *et al.* 1978). These studies further observed that

cadmium burdens in the common shrew *Sorex araneus*, a carnivorous species, were higher than both its dietary values and the cadmium burdens in two herbivorous species, *Apodemus sylvaticus* and *Microtus agrestis*. In contrast, lead and zinc showed no evidence of food chain accumulation. These findings indicate that cadmium displays a greater potential for accumulation through the food chain vegetation → invertebrate → predator than either lead or zinc.

Recently, feral pigeons *Columba livia* have been used as indicators of cadmium contamination in various parts of London (Hutton and Goodman 1980). Renal cadmium levels in populations from central London were moderately elevated and greater than those from suburban London, suggesting that contamination increases with proximity to the city centre. Markedly elevated cadmium levels were found in some birds from Heathrow Airport, indicating the presence of an appreciable degree of cadmium contamination at this site. The source of this hitherto unsuspected contamination is not clear at present, but aircraft may be implicated. The metal is used in their bodywork and there is a small cadmium plating works at the airport.

Apparently, there have been no reports of any cadmium-induced effects occurring in terrestrial vertebrates. This may in part be attributed to the absence of a sensitive and reliable bioanalytical index of cadmium exposure. This situation contrasts with that for lead where inhibition of ALA-D activity has been observed in several mammalian and avian species environmentally exposed to lead (Mouw *et al.* 1975; Finley, Dieter and Locke 1976; Hutton 1980).

### **3 Human exposure to cadmium**

#### **3.1 Introduction**

Dietary cadmium intake represents the single largest route of cadmium exposure in the general population. In smokers, however, cigarettes can be as important a source of cadmium as food. Additionally, children from contaminated localities may be exposed to elevated cadmium levels by the ingestion of cadmium-rich dusts or soils (see Section 3.4).

#### **3.2 Cadmium in diet and drink**

Dietary intake of cadmium has been measured by a number of Member States in the EC; Table 25 summarizes the values obtained from these

**Table 25 Average dietary intakes of cadmium in Member States of the EC**

| Country         | Daily Cd intake ( $\mu\text{g d}^{-1}$ ) | Methods of estimation            | Authority                   |
|-----------------|------------------------------------------|----------------------------------|-----------------------------|
| U.K.            | <18                                      | Standard diet                    | Hubbard and Lindsay (1979)  |
| F.R.G.          | 31                                       | Faecal analysis                  | Essing <i>et al.</i> (1969) |
| F.R.G.          | 48                                       | Standard diet                    | Essing <i>et al.</i> (1969) |
| France          | 20-30                                    | Standard diet/<br>Duplicate meal | CEC (1978)                  |
| Denmark         | <30                                      | Standard diet                    | Miljøministeriet (1980)     |
| The Netherlands | 29                                       | Standard diet                    | Ministry of Health (1980)   |
| U.S.A.          | 13-16                                    | Faecal analysis                  | Kowal <i>et al.</i> (1979)  |
| U.S.A.          | 26-61                                    | Standard diet                    | FDA (1977)                  |

Some estimates incorporate the contribution from water and beverages.

studies and also includes data from the U.S.A. Three methods have been used to estimate the dietary intakes of cadmium given in Table 25: the standard diet, the duplicate meal, and faecal analysis. The standard diet initially establishes a nationally representative diet by examining the eating habits of a typical population over a short time-period. The cadmium concentrations of the foodstuffs are then determined after preparation and cooking. The total dietary intake calculated is thus a statistical average and cannot reflect individual differences in eating habits. Duplicate diet studies involve the weighing and analysis of duplicate meals prepared at the same time as those consumed. Faecal excretion of cadmium can be used to estimate cadmium intake, after taking into account the proportion of ingested cadmium absorbed by the gut. This figure is generally assumed to be about 5 per cent.

Average dietary intake for the Member States shown in Table 25 varies from <18-48  $\mu\text{g d}^{-1}$ . The F.R.G. value of 48  $\mu\text{g}$  estimated by the standard diet method is significantly higher than that estimated by faecal analysis. Similarly, the intake measured for U.S.A. residents by faecal analysis is much lower than the FDA standard diet estimate. The reasons for the difference between the two methods of estimation are not clear. One explanation is that cadmium levels of individual foods are often lower than the analytical limits of detection, but such values are assumed to equal this limit when calculating the mean cadmium level of the food in question. Thus, the standard diet method may

overestimate the dietary intake of cadmium. Faecal cadmium levels, in contrast, are relatively high and present only minor analytical problems.

One point often ignored when comparing intake estimates from different populations or countries is that any estimate of dietary cadmium intake is strongly influenced by the quantity of food which is assumed to be eaten. For example, the average Danish dietary intake of  $<30 \mu\text{g d}^{-1}$  corresponds to a daily food consumption of 1.96 kg (Miljøministeriet 1980). U.K. food consumption is assumed to be somewhat lower at 1.47 kg daily (HMSO 1978); application of this figure to the Danish data would give a dietary intake of  $<22.5 \mu\text{g}$ , a similar value to the U.K. intake estimate.

In general, cadmium levels in drinking water are less than  $0.5 \mu\text{g l}^{-1}$  and intake from water and beverages is low compared with that from food. However, in view of the large quantity ( $\sim 2 \text{ l d}^{-1}$ ) of water consumed, even slight contamination would result in a significant increase in cadmium intake.

Table 26 shows the cadmium content of various food groups, the estimated mean intake of cadmium from each food group and total

**Table 26** Estimated average dietary intake of cadmium in the U.K. according to food class

| Food class           | Weight of food eaten ( $\text{kg d}^{-1}$ ) | Cadmium concentration ( $\mu\text{g g}^{-1}$ ) |                | Mean intake ( $\mu\text{g d}^{-1}$ ) |
|----------------------|---------------------------------------------|------------------------------------------------|----------------|--------------------------------------|
|                      |                                             | Mean*                                          | Range          |                                      |
| Cereals              | 0.23                                        | $<0.022$                                       | $<0.01-0.04$   | $<5$                                 |
| Meat                 | 0.15                                        | $<0.02$                                        | $<0.01-0.07$   | $<3$                                 |
| Fish                 | 0.02                                        | $<0.015$                                       | $<0.01-0.03$   | $<0.3$                               |
| Fats                 | 0.08                                        | $<0.012$                                       | $<0.01-0.03$   | $<1$                                 |
| Fruits and preserves | 0.17                                        | $<0.012$                                       | $<0.01-0.03$   | $<2$                                 |
| Root vegetables      | 0.18                                        | $<0.018$                                       | $<0.01-0.04$   | $<3$                                 |
| Other vegetables     | 0.11                                        | $<0.011$                                       | $<0.01-0.02$   | $<1$                                 |
| Milk                 | 0.40                                        | $<0.005$                                       | $<0.005-0.005$ | $<2$                                 |
| Beverages            | 0.12                                        | $<0.002$                                       | $<0.002-0.004$ | $<0.3$                               |
| Total                | 1.46                                        |                                                |                | $<18$                                |

Source: Lindsay (1980).

\* All values less than the limit of detection are assumed to equal the limit for purposes of calculating the mean.

dietary intake of the metal in the U.K. Cadmium is present at similar concentrations in all food groups. Thus, those foods consumed in the greatest amounts will make the largest dietary contribution to intake. In the case of U.K. consumers, cereals have been estimated to account for 30–40 per cent of the total dietary intake of cadmium (HMSO 1978).

Data generated from dietary intake studies in the EC are invariably presented as mean intakes of cadmium and as such give no indication of the frequency distribution of exposure for a population or a country. Thus it is not known what proportion of the EC population regularly exceed the maximum intake of 400–500  $\mu\text{g w}^{-1}$  recommended by the World Health Organization. Such information is needed in order to assess the extent of any hazard from cadmium exposure in the European Community. In the U.S.A., it has been claimed that as much as 15 per cent of the 15–20 year-old male population receives more than the WHO recommended maximum intake (Yost and Miles 1979). This figure is, however, considered to be an overestimate and is based upon relatively old analytical data of foodstuffs.

It is recommended that the distribution of dietary cadmium intakes for the general EC population should be determined and this should be done by the faecal excretion technique. This method, unlike the standard diet technique, would reflect individual variation in dietary habits and therefore cadmium intake. Ideally, such studies should continue for an extended period to obtain a clear estimate of the long-term exposure regime and eliminate any “noise” generated by short-term fluctuations in dietary intake.

Certain sections of the EC population are likely to be consuming higher than average quantities of dietary cadmium either because their diet is contaminated or because of unusual dietary habits. These critical consumers include people who eat large quantities of shellfish and those who eat vegetables grown on contaminated soils. Such soils include those which have received heavy applications of sewage sludge, those in areas of mineralization, or those in the vicinity of lead–zinc mines and point sources of cadmium (Table 23). It should be emphasized that even individuals from these contaminated localities will have to consistently consume unusually large quantities of home-grown vegetables before they exceed the WHO provisional tolerable weekly intake on a regular basis.

The average absorption of cadmium from the gut is generally assumed to be about 4–6 per cent of the total ingested. This average value,

however, masks the existence of a large individual variation in uptake. For example, one uptake study using only 14 subjects reported an absorption range of some 0.7–14.6 per cent (McLellan, Flanagan, Chamberlain and Valberg 1978). This large variation suggests that a proportion of the general population will display a significantly higher absorption of ingested cadmium than the average.

Several nutritional factors can influence the uptake of ingested cadmium. A low dietary intake of calcium, for example, results in an enhanced absorption of cadmium. Of more importance is the fact that individuals with moderate iron deficiency have been shown to have significantly higher absorption rates than subjects with normal iron stores (Flanagan *et al.* 1978). A slight iron deficiency is not an uncommon disorder, particularly in females, and this should be taken into account when estimating the likely range of absorption rates in a particular population.

### 3.3 Inhalation and smoking

It has been estimated that 20–30 per cent of the airborne cadmium inhaled by humans is deposited in the pulmonary region of the lung, where some 65 per cent is absorbed (Friberg *et al.* 1974; CEC 1978). Thus, between 13 and 20 per cent of the cadmium in air will be absorbed. It is unusual for airborne cadmium levels regularly to exceed  $20 \text{ ng m}^{-3}$ ; an adult male with a respiratory volume of  $15 \text{ m}^3 \text{ d}^{-1}$  is therefore unlikely to inhale more than  $0.3 \text{ } \mu\text{g}$  daily and absorb  $0.04\text{--}0.06 \text{ } \mu\text{g}$  cadmium.

Populations living very close to point sources of airborne cadmium will be exposed to larger quantities of the metal. At an ambient level of  $1 \text{ } \mu\text{g m}^{-3}$  for example, an adult male may absorb  $2\text{--}3 \text{ } \mu\text{g}$  cadmium daily, a quantity equivalent to an additional dietary intake of about  $50 \text{ } \mu\text{g d}^{-1}$ . It must be stressed that this calculation ignores the large day-to-day fluctuations in airborne cadmium levels which occur around point sources. Additionally, the unrealistic assumption is made that residents of such areas spend 24 hours of any day close to the emission source.

Tobacco contains relatively high cadmium concentrations and cigarettes represent an important source of exposure for smokers. Cadmium is absorbed relatively efficiently from cigarette smoke; one study has calculated that the consumption of 20 cigarettes results in the absorption of  $1.4 \text{ } \mu\text{g}$  cadmium (EPA 1979). This quantity of cadmium is equivalent

to the daily ingestion of an additional 25–30  $\mu\text{g}$  dietary cadmium. Obviously, those individuals who regularly smoke two or more packets of cigarettes will absorb correspondingly greater quantities of cadmium.

### 3.4 Ingestion of cadmium-rich dusts and soils

Some young children regularly ingest small quantities of dusts and soils. It is thus conceivable that this habit may result in a significant uptake of cadmium in contaminated localities. The average daily intake of dirt via the hands in young children has been estimated to be 100 mg (Lepow *et al.* 1974). Application of this figure to the household dust concentrations of 193  $\mu\text{g g}^{-1}$  found near to a lead refinery in the U.K. (Muskett *et al.* 1979) suggests that children in the area will daily ingest some 20  $\mu\text{g}$  cadmium. This is equivalent to the doubling of dietary intake for such children.

### 3.5 Total intake and absorption and critical levels of cadmium exposure

Table 27 summarizes the range of cadmium intakes and levels of absorption likely to occur in most adults from the EC. The major variables affecting net retention are the two levels of dietary intake (20 and 60  $\mu\text{g d}^{-1}$ ), two absorption rates (5 and 10 per cent) and differing patterns of cigarette consumption (0–2 packets daily). Overall, retention of cadmium ranges from 1.05  $\mu\text{g}$  to 8.85  $\mu\text{g}$ . Heavy smokers and individuals with slight iron deficiency or elevated dietary intake will absorb considerably more cadmium.

The values given in the last column of Table 27 may be compared with the amount of cadmium needed to be absorbed daily to bring about renal dysfunction after 50 years. Lauwerys (1981) has estimated that this 50-year critical retention value is between 10 and 15  $\mu\text{g}$ . In contrast, some work by Kjellström indicates that as little as 5–6  $\mu\text{g}$  cadmium retained daily over a 50-year period will result in renal tubular dysfunction in about one per cent of the population (Kjellström 1980). The dose-response model devised by Kjellström was, however, based on limited populations from Japan consisting mainly of elderly women. Dietary intake of cadmium may not have been assessed accurately and certain factors in addition to cadmium may have contributed to the development of proteinuria.

**Table 27 Total daily absorption of cadmium and relative contributions from the major sources of exposure**

| Intake<br>( $\mu\text{g d}^{-1}$ ) | Diet and water         |                                 | Respiratory sources                                                   |                                                  | Total<br>absorption<br>( $\mu\text{g}$ ) |
|------------------------------------|------------------------|---------------------------------|-----------------------------------------------------------------------|--------------------------------------------------|------------------------------------------|
|                                    | Absorption<br>rate (%) | Absorption<br>( $\mu\text{g}$ ) | Air Cd at<br>20 $\text{ng m}^{-3}$<br>absorption<br>( $\mu\text{g}$ ) | Smoking<br>status-<br>packets<br>$\text{d}^{-1}$ |                                          |
| 20                                 | 5                      | 1                               | 0.05                                                                  | 0 0                                              | 1.05                                     |
|                                    |                        |                                 |                                                                       | 1 1.4                                            | 2.45                                     |
|                                    |                        |                                 |                                                                       | 2 2.8                                            | 3.85                                     |
|                                    | 10                     | 2                               | 0.05                                                                  | 0 0                                              | 2.05                                     |
|                                    |                        |                                 |                                                                       | 1 1.4                                            | 3.45                                     |
|                                    |                        |                                 |                                                                       | 2 2.8                                            | 4.85                                     |
| 60                                 | 5                      | 3                               | 0.05                                                                  | 0 0                                              | 3.05                                     |
|                                    |                        |                                 |                                                                       | 1 1.4                                            | 4.45                                     |
|                                    |                        |                                 |                                                                       | 2 2.8                                            | 5.85                                     |
|                                    | 10                     | 6                               | 0.05                                                                  | 0 0                                              | 6.05                                     |
|                                    |                        |                                 |                                                                       | 1 1.4                                            | 7.45                                     |
|                                    |                        |                                 |                                                                       | 2 2.8                                            | 8.85                                     |

For the present, it will be assumed that the lower figure given by Lauwerys (1981),  $10 \mu\text{g d}^{-1}$ , is the 50-year critical retention value. The range of retention values shown in Table 27 indicate that according to the figure chosen, individuals within the EC will display a safety margin of 1–10. Thus a proportion of the EC population, probably a large majority, cannot be considered to be at risk from cadmium exposure. On the other hand, a small fraction of the population will be daily absorbing quantities of cadmium as large as  $8.85 \mu\text{g}$  and such individuals must be considered to be at risk from cadmium exposure.

#### 4 Future trends in dietary cadmium intake in the EC

##### 4.1 Introduction

The previous section examined present-day exposure to cadmium in the general population of the EC. Attention will now be directed at the

consequences of long-term cadmium inputs to agricultural land upon dietary intakes of the metal.

The consumption of plant-based foodstuffs represents the single largest source of dietary cadmium intake for the general population (Table 26). For this reason, predictions of future changes in the cadmium content of plants will focus upon the situation for plant-based foods, particularly cereal crops and root vegetables. Initially, forecasts will be made for future soil and plant cadmium concentrations, together with the corresponding dietary intakes. This will be followed by an attempt to predict the effectiveness of adopting various control strategies which reduce the inputs of cadmium to agricultural soils.

In the absence of relevant information, several simplifications and assumptions have to be made in studies of this nature. For example, it is not possible to predict the behaviour of cadmium in the soil-plant system for all the soil types encountered in the European Community. However, some insight into the influence of soil type upon the behaviour of cadmium has been gained by utilizing two adsorption coefficients (see Section 2.5.1) thought to cover the range of values found in most soils.

One factor which was not evaluated but may be of importance in terms of an additional contribution to dietary intake is the possibility of post-harvest contamination of plant-based foods. It is conceivable that processing and packaging represent a significant source of cadmium contamination but this will be difficult to quantify. An additional assumption which has been made in this study is that all the plant-based foods consumed in the European Community are produced from crops grown in the region. This is, of course, an oversimplification; in the U.K. for example, nearly all the wheat used to make bread and other wheat-based products actually comes from the U.S.A.

#### 4.2 Characterization of the behaviour of cadmium in soil-plant systems

The transport model used to predict the future behaviour of cadmium in soil-crop plant systems is based upon the exposure commitment approach which has been developed at MARC. The relevant reservoirs of cadmium and their transfer sequence are shown in Figure 1. The various fluxes through the system are described by transfer coefficients, defined as the ratio of the cadmium concentrations in the receptor and donor compartments.

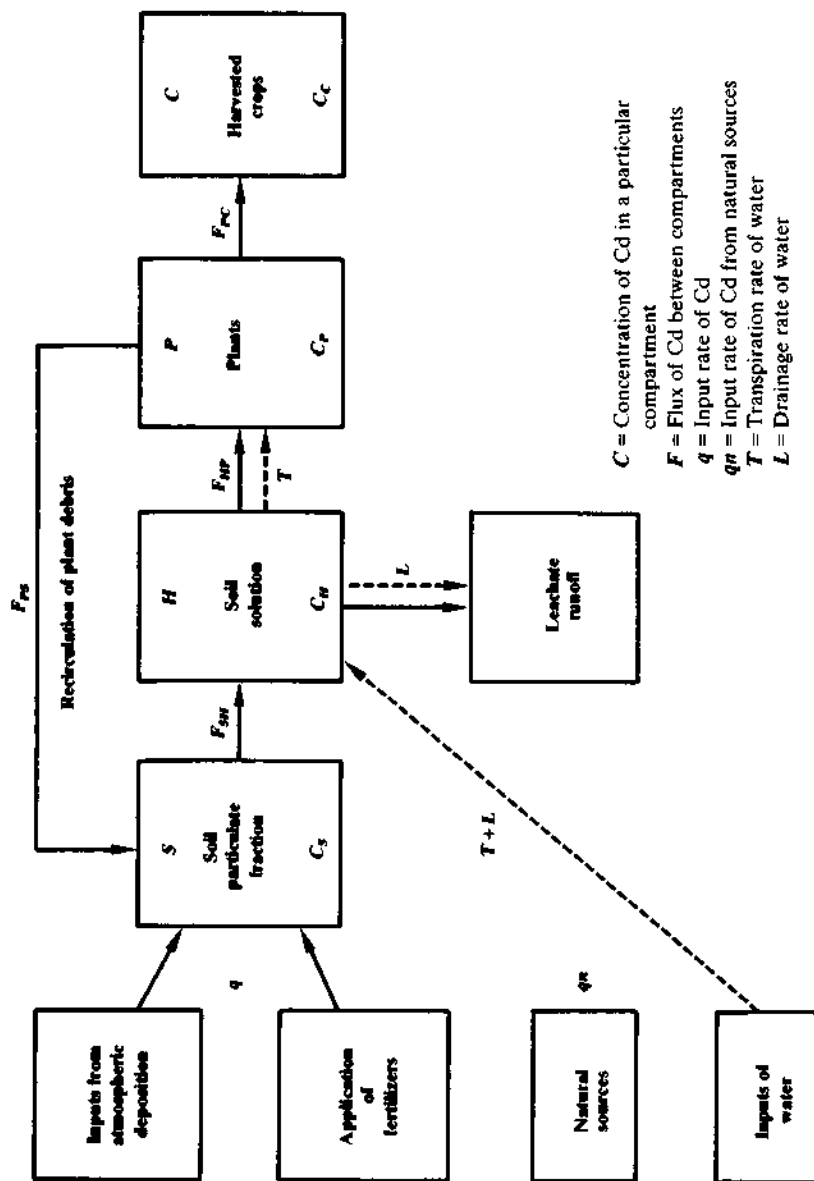


Figure 1 Soil-plant transfer pathways of cadmium in background agricultural areas of the EC

The importance of the adsorption coefficient,  $\alpha$ , as a determinant of the behaviour of cadmium in the soil system has been discussed briefly in Section 2.5.1. Two values of  $\alpha$ , considered to cover the range commonly found in soils from the EC, were calculated from a study of cadmium sorption in nine agricultural soils from the U.K. (Jarvis and Jones 1980). There was a threefold range in the two extremes of  $\alpha$  in these soils,  $6.76 \times 10^{-3}$  and  $18.2 \times 10^{-3} \text{ g ml}^{-1}$ .

The selectivity coefficient,  $S$ , has been used to characterize cadmium uptake by cereal crops. As discussed in Section 2.5.1, the selectivity coefficient can vary widely between different plant species. For this reason, two values of  $S$  were used, 0.1 and 1. These are considered to encompass the range encountered in plant species and are similar to the values used in another soil-plant cadmium study by Poelstra *et al.* (1979).

Other parameters employed include the water flow associated with plant production,  $\beta$ . The rate of plant growth determines the cadmium content of the plant, corresponding to a given water uptake. For the sake of simplicity, it has been assumed that the transpiration rate and the plant production rate are related by the constant of proportionality,  $\beta$ . Additionally, it is necessary to specify the effective soil depth,  $d$ , the soil density  $\sigma$ ; the soil moisture content  $\theta$ , the transpiration rate  $T$  and the soil leaching rate  $L$ . Finally, the recirculation factor  $\gamma$  is the fraction of cadmium in plants which is recycled to soils after harvest at the end of each season. In cereals,  $\gamma$  is high and a value of 0.8 has been assumed

**Table 28** Parameters used to characterize the behaviour of cadmium in soil-plant systems

| Parameter                                   | Symbol   | Chosen value                                                                               | Units                                    |
|---------------------------------------------|----------|--------------------------------------------------------------------------------------------|------------------------------------------|
| Adsorption coefficient                      | $\alpha$ | $\left\{ \begin{array}{l} 6.76 \times 10^{-3} \\ 18.2 \times 10^{-3} \end{array} \right\}$ | $\text{g ml}^{-1}$                       |
| Selectivity coefficient                     | $S$      | 0.1, 1                                                                                     | —                                        |
| Transpiration rate (water)                  | $T$      | 20                                                                                         | $\text{ml cm}^{-2} \text{ y}^{-1}$       |
| Drainage rate (water)                       | $L$      | 20                                                                                         | $\text{ml cm}^{-2} \text{ y}^{-1}$       |
| Soil density                                | $\sigma$ | 1.3                                                                                        | $\text{g cm}^{-3}$                       |
| Soil depth                                  | $d$      | 30                                                                                         | cm                                       |
| Soil moisture content                       | $\theta$ | 0.25                                                                                       | $\text{ml cm}^{-3}$<br>(dimensionless)   |
| Water flow associated with plant production | $\beta$  | 200                                                                                        | $\text{ml y}^{-1}$ per $\text{g y}^{-1}$ |
| Fraction of plant Cd recycled to soil       | $\gamma$ | 0.8                                                                                        | —                                        |

for this study. A list of these parameters, their symbols, units and chosen values is given in Table 28.

The behaviour of cadmium in soil-plant systems can now be determined by the use of the following coefficients:

### 1. The residence time of cadmium in soil $G_S$

This coefficient determines the steady-state concentration of cadmium in soils for a given, constant and continuing input rate of cadmium,  $q$ . It also determines how long it will take for the steady state to be reached. The longer the residence time, the higher the ultimate steady-state concentration of cadmium in soil.

The mean residence time is defined by

$$G_S = \frac{(\sigma + \theta\alpha)d}{\alpha(ST(1 - \gamma) + L)}$$

Thus the residence time is inversely proportional to the adsorption coefficient,  $\alpha$ .

The steady-state concentration of cadmium in soils  $C_S^*$  can be calculated by multiplying the residence time,  $G_S$  with the input rate,  $q$ . Thus, if the input rate of cadmium is  $2 \text{ g ha}^{-1} \text{ y}^{-1}$  and the residence time is 300 years the steady-state concentration is  $600 \text{ g ha}^{-1}$ .

The transient time series, starting from zero contamination is given by:

$$\begin{aligned} C_S(t) &= C_S^* (1 - \exp - t/G_S) \\ &= qG_S(1 - \exp - t/G_S) \end{aligned}$$

### 2. The soil-plant transfer coefficient $P_{SP}$

This coefficient simply defines the relationship between cadmium concentrations in plants and soils,  $C_S$  and  $C_P$ .

$$\text{i.e. } P_{SP} = \frac{C_P}{C_S}$$

The formula for this coefficient will vary according to the manner in which soil concentration is expressed ( $\mu\text{g g}^{-1}$  or  $\text{g ha}^{-1}$ ). When soil and plant cadmium are expressed in  $\mu\text{g g}^{-1}$ , the expression is

$$P_{SP} = \frac{C_P}{C_S} = \frac{\beta S \alpha \sigma}{(\sigma + \theta \alpha)}$$

When soil cadmium is expressed per unit surface area, for a given depth, the expression is

$$P_{SP} = \frac{\beta S \alpha}{d(\sigma + \theta \alpha)}$$

$P_{SP}$  is proportional to the product of  $S$  and  $\alpha$  because  $\sigma \gg \theta \alpha$ .

### 3. The source to plant transfer coefficient $sP_{OP}$

This coefficient defines the relationship between the input rate of cadmium to soils,  $q$ , and the eventual concentration in plants once equilibrium has been reached in soils,

$$\text{i.e. } sP_{OP} = \frac{C_P^*}{q}$$

The formula for this coefficient is

$$sP_{OP} = \frac{\beta S}{ST(1 - \gamma) + L}$$

In transient conditions, the plant cadmium concentration,  $C_P(t)$ , will follow the concentration in soil,  $C_S(t)$ . The formula for  $C_P(t)$  is

$$C_P(t) = C_P^* (1 - \exp - t/G_S) = q sP_{OP} (1 - \exp - t/G_S)$$

The detailed derivations of these formulae are given in Appendix 7.

## 4.3 Inputs of cadmium to arable soils in the EC

The total cadmium input to arable soils in areas away from localized contamination is assumed to be  $8 \text{ g ha}^{-1} \text{ y}^{-1}$ . Phosphate fertilizer application is estimated to contribute  $5 \text{ g ha}^{-1} \text{ y}^{-1}$  (see Table 21) while atmospheric deposition accounts for the remaining  $3 \text{ g ha}^{-1} \text{ y}^{-1}$  (see Section 2.3). The contribution from sewage sludge applications is considered to be too small on a national or regional basis to warrant inclusion. A study predicting cadmium concentrations in Danish soils employed the lower total of  $5.1 \text{ g ha}^{-1} \text{ y}^{-1}$  inputs to background soils (Tjell, Hansen, Christensen and Hovmand, 1981). This figure was made up of

3 g ha<sup>-1</sup> y<sup>-1</sup> from fertilizers, 2 g ha<sup>-1</sup> y<sup>-1</sup> from atmospheric deposition and just 0.1 g ha<sup>-1</sup> y<sup>-1</sup> from sewage sludge.

The natural source component,  $q_n$ , has not been quantified. Instead, it has been assumed that soil and plant concentrations of cadmium have already reached equilibrium from this source and that natural inputs account for a burden in soil of 0.66 kg ha<sup>-1</sup>, corresponding to 0.17 µg g<sup>-1</sup> (see Appendix 7).

#### 4.4 Predicted changes in soil and plant cadmium concentrations and influence upon dietary intake

The transfer coefficients used to predict soil and plant cadmium concentrations are shown in Table 29. The four scenarios of soil and plant cadmium, depicted in Table 30, were calculated after assuming a present-day soil cadmium content of 0.78 kg ha<sup>-1</sup> (0.2 µg g<sup>-1</sup>). Both soil and plant cadmium levels have been calculated for 100 years from now and for the steady state. Additionally, plant cadmium levels were computed for the present day and also for the concentration associated with natural sources. The predicted accumulation of soil cadmium increases with residence time. It should be noted, however, that the highest plant cadmium concentrations are associated with the fourth scenario, where soil cadmium levels are lowest.

The large range of values predicted for future plant cadmium levels in Table 30 emphasizes the problems involved in estimating overall changes in dietary intake of the metal. Of the four scenarios, the first is probably the most representative of the future situation in the EC. The adsorption coefficient used in the first and second scenarios is the

**Table 29 Cadmium transfer coefficient estimates for soil-plant systems with differing values of adsorption coefficient and selectivity coefficient**

| Scenario | $\alpha$              | $S$ | Soil residence time (y) | Soil-plant transfer coefficient (µg g <sup>-1</sup> per kg ha <sup>-1</sup> ) | Source-plant transfer coefficient (µg g <sup>-1</sup> per kg ha <sup>-1</sup> y <sup>-1</sup> ) |
|----------|-----------------------|-----|-------------------------|-------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------|
| 1        | $6.76 \times 10^{-3}$ | 0.1 | 283                     | 0.035                                                                         | 9.8                                                                                             |
| 2        | $6.76 \times 10^{-3}$ | 1   | 241                     | 0.35                                                                          | 83                                                                                              |
| 3        | $18.2 \times 10^{-3}$ | 0.1 | 106                     | 0.093                                                                         | 9.8                                                                                             |
| 4        | $18.2 \times 10^{-3}$ | 1   | 90                      | 0.93                                                                          | 83                                                                                              |

**Table 30 Four scenarios of soil and plant cadmium concentrations in the EC**

| Scenario | Soil concentrations<br>(kg ha <sup>-1</sup> ) |         | Plant concentrations<br>(µg g <sup>-1</sup> ) |         |             |         |
|----------|-----------------------------------------------|---------|-----------------------------------------------|---------|-------------|---------|
|          | $C_s^{100}$                                   | $C_s^*$ | $C_p^n$                                       | $C_p^0$ | $C_p^{100}$ | $C_p^*$ |
| 1        | 1.42                                          | 2.92    | 0.023                                         | 0.027   | 0.050       | 0.102   |
| 2        | 1.395                                         | 2.59    | 0.23                                          | 0.273   | 0.488       | 0.907   |
| 3        | 1.23                                          | 1.51    | 0.061                                         | 0.073   | 0.114       | 0.140   |
| 4        | 1.18                                          | 1.38    | 0.61                                          | 0.73    | 1.097       | 1.28    |

$C_s^{100}$  = soil cadmium concentration after 100 years.

$C_s^*$  = steady-state soil cadmium concentration.

$C_p^n$  = plant cadmium concentration associated with natural sources.

$C_p^0$  = present plant cadmium concentration.

$C_p^{100}$  = plant cadmium concentration after 100 years.

$C_p^*$  = steady-state plant cadmium concentration.

lower of the two values and is likely to be nearest to the values found in most agricultural soils. Similarly, the lower selectivity coefficient values employed in scenarios one and three is thought to be the more appropriate for cereal crops and root vegetables. Scenarios two and four indicate that present-day plant cadmium concentrations are 0.27 µg g<sup>-1</sup> and 0.73 µg g<sup>-1</sup> respectively; both values are unrealistically high for cereal crops and most vegetables of dietary importance.

Present-day plant concentrations of cadmium computed from scenario one (0.027 µg g<sup>-1</sup>) fall within the range actually found in cereals and root vegetables. In the U.K., it has been estimated that a total of 410 g cereal products and root vegetables is consumed daily (Table 26). Application of this quantity to the present-day hypothetical plant concentration (Table 30) results in a daily intake of 11 µg cadmium from these foodstuffs. The actual intake of cadmium from these sources in the U.K. has been given by Lindsay (1980) as <8 µg. For the purposes of this study, it is assumed that the contribution from all other food classes to daily dietary intake in the U.K. is 10 µg. Thus, the hypothetical present-day dietary intake of cadmium is assumed to be 21 µg. Plant cadmium concentrations 100 years in the future are forecast to be nearly double those of the present day, at 0.050 µg g<sup>-1</sup>. The corresponding dietary intake from cereal products and root vegetables is calculated to increase to 20.5 µg d<sup>-1</sup>. The contribution from other food classes such as dairy products, meat and fish, is unlikely to be significantly affected

by increased soil cadmium concentrations and dietary intake is therefore predicted to increase to  $30.5 \mu\text{g d}^{-1}$  by the year 2080. The situation in Denmark may be more critical, as the daily consumption of cereal products and vegetables is greater than in the U.K. and totals 591 g (Miljøministeriet 1980). Thus, cadmium intake from these food classes 100 years hence is predicted to be  $29.6 \mu\text{g d}^{-1}$  ( $591 \text{ g} \times 0.05 \mu\text{g g}^{-1}$ ). The daily cadmium contribution from other food classes is estimated to be 12  $\mu\text{g}$  (Miljøministeriet 1980), indicating that future dietary intake will be about  $42 \mu\text{g d}^{-1}$  in Denmark.

The predictions of dietary intake for the U.K. and Denmark are summarized in Table 31. Steady-state values are also given. The soil properties associated with scenario three are likely to be found in a number of areas in Europe. For this reason, future dietary intakes associated with this scenario are also given in Table 31. Those intakes are larger than predicted by scenario one, with 100-year values of  $57 \mu\text{g d}^{-1}$  assigned to the U.K. and  $79 \mu\text{g d}^{-1}$  to Denmark. It should also be noted that scenario three plant concentrations as little as 10 years in the future are  $0.079 \mu\text{g g}^{-1}$ —a value actually higher than the 100-year value for scenario one.

In summary, predictions of daily dietary intake in the EC for 100 years in the future range from  $30.5$ – $79 \mu\text{g}$  cadmium. These estimates vary according to the assumed consumption of vegetables and cereal products and also the properties of the soil from which the plants were grown.

The predicted intake values given above are significantly higher than those encountered in the EC at present. Accompanying these future average intakes will be a larger proportion of the population who exceed the tolerable intake recommended by the WHO. In view of the implications of these predictions, it is suggested that both the modelling tech-

**Table 31** Present and future dietary intakes ( $\mu\text{g g}^{-1}$ ) in the U.K. and Denmark for two scenarios of soil and plant cadmium concentrations

| Country | Present-day intake | Scenario 1      |              | Scenario 3      |              |
|---------|--------------------|-----------------|--------------|-----------------|--------------|
|         |                    | 100 years hence | Steady state | 100 years hence | Steady state |
| U.K.    | ~21                | 30.5            | 52           | 57              | 67           |
| Denmark | ~30                | 42              | 72           | 79              | 95           |

nique and the values used in the model be subject to further validation. Until this is done, the forecasts given above can only be viewed as possible indications of the future situation.

#### 4.5 Impact of control strategies on future dietary intake of cadmium

In recent years, concern has been expressed that human exposure to cadmium is unacceptably high and it has been suggested that steps should be taken to reduce future intake of the metal. This concern has led to proposals for restricting or even banning the use of certain cadmium-containing materials. These proposals have not, however, been critically examined to assess whether action of this nature will actually result in a significant reduction of dietary intake. For this reason, the impact of two hypothetical control strategies upon future soil and plant cadmium levels in the EC will be assessed. It must be emphasized that the techniques developed in this study only allow predictions of future soil, plant and dietary intake levels on an average basis. Local problems of soil contamination arising from sewage sludge applications or airborne deposition in the proximity of point sources may be of great importance in many areas. It has not been possible, however, to assess the influence of such contamination on the dietary intake of cadmium at a national level.

##### *1. Removal of the cadmium input associated with phosphate fertilizer applications*

The application of phosphate fertilizers results in the single largest cadmium input to agricultural soils and has been estimated to amount to  $5 \text{ g ha}^{-1} \text{ y}^{-1}$ . The removal of this input may, in theory at least, be brought about by a calcining process or by the use of phosphate rock which contains negligible amounts of cadmium.

It is forecast that removal of cadmium inputs from phosphate fertilizers would produce the following plant cadmium concentrations:

|                                 |                              |
|---------------------------------|------------------------------|
| after 10 years, scenario one    | $= 0.028 \mu\text{g g}^{-1}$ |
| after 100 years, scenario one   | $= 0.035 \mu\text{g g}^{-1}$ |
| after 10 years, scenario three  | $= 0.074 \mu\text{g g}^{-1}$ |
| after 100 years, scenario three | $= 0.084 \mu\text{g g}^{-1}$ |

It is thus predicted that this control strategy would result in much smaller increases in future plant cadmium concentrations. For scenario one, the

100-year forecasts of total dietary intake associated with this control strategy are only  $24 \mu\text{g d}^{-1}$  for the U.K. and  $33 \mu\text{g d}^{-1}$  for Denmark.

## 2. The cessation of atmospheric cadmium emissions from the iron and steel industries

It was estimated earlier that iron and steel production in the EC is the single largest source of airborne cadmium in the EC and represents 26 per cent of the total emissions (Table 16). For the purposes of this study, the simplistic assumption will be made that emissions from this source also account for 26 per cent of the background atmospheric deposition rate of  $3 \text{ g ha}^{-1} \text{ y}^{-1}$ . Thus, the abolition of airborne cadmium emissions from this source would reduce atmospheric deposition to  $2.2 \text{ g ha}^{-1} \text{ y}^{-1}$ . Such a control strategy would necessitate the banning of cadmium-plated steel or its exclusion from recycled steel scrap.

The implementation of this control strategy is predicted to produce the following plant cadmium concentrations:

|                                 |                              |
|---------------------------------|------------------------------|
| after 10 years, scenario one    | $= 0.03 \mu\text{g g}^{-1}$  |
| after 100 years, scenario one   | $= 0.047 \mu\text{g g}^{-1}$ |
| after 10 years, scenario three  | $= 0.078 \mu\text{g g}^{-1}$ |
| after 100 years, scenario three | $= 0.109 \mu\text{g g}^{-1}$ |

Comparison of these values with those in Table 30 indicate that the abolition of the single largest source of airborne cadmium in the EC would have minimal influence upon future plant cadmium levels. Similarly, dietary intake of cadmium would increase at only a slightly lower rate than that forecast in the absence of this control strategy.

Again, it must be made clear that further validation of the model is required before the above findings can be considered as anything more than indications of the impact of control strategies on future cadmium intake.

## References

- 1 Andersen, C. 1979 Cadmium, lead and calcium content, number and biomass, in earthworms (*Lumbricidae*) from sewage sludge treated soil. *Pedobiologia* **19**, 309–319.
- 2 Archer, F. C. 1980 Trace elements in soils in England and Wales. In: *Inorganic Pollution and Agriculture*, HMSO, London, 184–190.
- 3 Badanur, V. P. and Venkata Rao, B. V. 1972 Influence of phosphorus buildup on availability of micronutrients in red soils of Bangalore. *Soil Sci.* **116**, 292–294.

- 4 Beauford, W., Barber, J. and Barringer, A. R. 1977 Release of particles containing metals from vegetation into the atmosphere. *Science* **195**, 571-573.
- 5 Belot, Y., Diop, B. and Marini, T. 1971 Composition minérale de la matière particulaire en suspension dans l'air dans des zones urbaines. Cited in Rahn (1976).
- 6 Berrow, M. L. and Burridge, J. C. 1979 Sources and distribution of trace elements in soils and related crops. In: *Management and Control of Heavy Metals in the Environment*, London, September 1979, CEP Consultants Ltd, Edinburgh, 304-311.
- 7 Bolton, N., Van Hook, R., Fulkerson, W., Lyon, W. and Andren, A. 1973 *Trace element measurements at the coal-fired Allen Steam Plant*. ORNL-NSF-EP-43. Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- 8 Branson, J. and Pattenden, N. J. 1979 *Multi-element airborne dust measurements for Borough of Lambeth. Report for year April 1978 to March 1979*. AERE Harwell Report G-1526, Harwell, Oxon.
- 9 Bryan, G. W. 1973 The occurrence and seasonal variation of trace metals in the scallops *Pecten maximus* (L.) and *Chlamys opercularis* (L.). *J. Mar. Biol. Assoc. U.K.* **53**, 145-166.
- 10 Buat-Ménard, P. and Arnold, M. 1978 The heavy metal chemistry of atmospheric particulate matter emitted by Mount Etna volcano. *Geophys. Res. Lett.* **5**, 245-248.
- 11 Burkitt, A., Lester, P. and Nickless, G. 1972 Distribution of heavy metals in the vicinity of an industrial complex. *Nature* **238**, 327-328.
- 12 Cataldo, D. A. and Wildung, R. E. 1978 Soil and plant factors influencing the accumulation of heavy metals in plants. *Environ. Health Perspect.* **27**, 149-159.
- 13 Cawse, P. A. 1977 *A Survey of atmospheric trace elements in the U.K.: results for 1976*. AERE Harwell Report R-8869, HMSO, London.
- 14 Cawse, P. A. 1979 *Trace elements in soil, vegetables and rainwater in Walsall*. AERE Harwell Report G-1343, Harwell, Oxon.
- 15 CEC 1978 *Criteria (Dose/Effect Relationships) for Cadmium*. Pergamon Press, Oxford.
- 16 CEC 1979 *Raw materials. Research and development. I. Copper, II. Lead and Zinc*. Commission of the European Communities, DGXII, Brussels.
- 17 Chatterjee, P. K. and Pooley, F. D. 1977 An examination of some trace elements in South Wales coals. *Proc. Australas. Inst. Min. Metall.* **263**, 19-30.
- 18 Coleman, R. et al. 1979 *Sources of atmospheric cadmium* EPA-450/5-79-006, U.S. Environmental Protection Agency.
- 19 Cunningham, L. M., Collins, F. W. and Hutchinson, T. C. 1975 Physiological and biochemical aspects of cadmium toxicity in soybean. 1. Toxicity symptoms and autoradiographic distribution of Cd in roots, stem and leaves. In: *International Conference on Heavy Metals in the Environment*, Toronto, Canada, 27-31 October 1975, 97-120.
- 20 Davis, R. D. and Coker, E. G. 1980 *Cadmium in Agriculture, with Special Reference to the Utilization of Sewage Sludge on Land*. Water Research Centre, Report TR 139, Stevenage, Herts.
- 21 Deane, G. L., Lynn, D. A. and Surprenant, N. F. 1976 *Cadmium: Control Strategy Analysis*, GCA-TR-75-36-G, U.S. Environmental Protection Agency.
- 22 DOE 1980 *Cadmium in the environment and its significance to man*. Department of the Environment, Pollution Paper No. 17, HMSO, London.

- 23 Dugdale, P. J. and Hummel, B. L. 1978 Cadmium in the lead smelter at Belledune: its association with heavy metals in the environment. In: *First International Cadmium Conference San Francisco*, January 1977, Metal Bulletin Ltd, 53-75.
- 24 Duggan, M. J. 1980 Lead in urban dust: an assessment. *Water, Air, Soil, Pollut.* **14**, 309-321.
- 25 EPA 1979 *Health Assessment Document for Cadmium* EPA-600/8-79-003 Environmental Protection Agency, U.S.A.
- 26 Essing, H.-G., Schaller, K.-H., Szadkowski, D. and Lehnert, G. 1969 Usuelle cadmiumbelastung durch Nahrungsmittel und Getränke. *Arch. Hyg. Bikt.* **153**, 490-494.
- 27 Eurostat 1980 *Energy statistics yearbook 1978*. Statistical Office of the European Communities, Commission of the European Communities, Brussels.
- 28 Feenstra, J. F. 1978 *Control of water pollution due to cadmium discharged in the process of producing and using phosphate fertilizers*. CEC Report: ENV/223/74-EN, Commission of the European Communities, Brussels.
- 29 FDA 1977 *Compliance Program Evaluation, FY 1974 Total Diet Studies (7320.08)*. FDA Bureau of Foods, Washington D.C.
- 30 Finley, M. T., Dieter, M. P. and Locke, L. N. 1976  $\delta$ -aminolevulinic acid dehydratase: inhibition in ducks dosed with lead shot. *Environ. Res.* **12**, 243-249.
- 31 Flanagan, P. R., McLellan, J. S., Haist, J., Cherian, G., Chamberlain, H. J. and Valberg, L. S. 1978 Increased dietary cadmium absorption in mice and human subjects with iron deficiency. *Gastroenterology* **74**, 841-846.
- 32 Friberg, L., Piscator, M., Nordberg, G. and Kjellström, T. 1974 *Cadmium in the environment*, 2nd edition, CRC Press, Cleveland, Ohio.
- 33 Gibson, J. 1979 *Coal and the environment*. National Coal Board, London.
- 34 Gingell, S. M., Campbell, R. and Martin, M. H. 1976 The effect of zinc, lead and cadmium pollution on the leaf surface microflora. *Environ. Pollut.* **11**, 25-37.
- 35 Gish, C. D. and Christensen, R. E. 1973 Cadmium, nickel, lead and zinc in earthworms from roadside soils. *Environ. Sci. Technol.* **11**, 1060-1063.
- 36 Greenberg, R. R., Zoller, W. H. and Gordon, G. E. 1978 Composition and size distribution of particles released in refuse incineration. *Environ. Sci. Technol.* **12**, 566-573.
- 37 Harrison, R. M. 1979 Toxic metals in street and household dusts. *Sci. Total Environ.* **11**, 89-97.
- 38 Harrison, R. M., Perry, R. and Wellings, R. A. 1975 Lead and cadmium in precipitation: their contribution to pollution. *J. Air Pollut. Control Assoc.* **25**, 627-630.
- 39 Heinrichs, H. 1977 Emissions of 22 elements from brown coal combustion. *Naturwiss.* **64**, 479-481.
- 40 Hiscock, S. A. 1978 *Production, consumption and uses of cadmium in the European Community (1965-1976)*. ENV/223/74-E. Commission of the European Communities, Brussels.
- 41 HMSO 1978 *The surveillance of food contamination in the United Kingdom*. Food surveillance paper No. 1.
- 42 Hofstader, R. A., Milner, O. I. and Runnels, J. H. (Eds) 1976 Cadmium. In: *Analysis of Petroleum for Trace Metals; Advances in Chemistry Series: 156*. American Chemical Society, 81-94.

- 43 Holmes, R. 1975 The regional distribution of cadmium in England and Wales. PhD Thesis, University of London.
- 44 Horler, D. N. N. and Barbour, J. 1979 Relationships between vegetation and heavy metals in the atmosphere. In: *Management and Control of Heavy Metals in the Environment*, London, September 1979, CEP Consultants Ltd, Edinburgh, 275-278.
- 45 Hovmand, M. F. 1980 Cycling of lead, cadmium, copper, zinc and nickel in Danish agriculture. In: *Land Application of Wastewater Sludge*. Vol. II, Chapter IV, PF Forlag, Lyngby, Denmark.
- 46 Hubbard, A. W. and Lindsay, D. G. 1979 Dietary intakes of heavy metals by consumers in the United Kingdom. In: *Management and Control of Heavy Metals in the Environment*, London, September 1979, CEP Consultants Ltd, Edinburgh, 52-55.
- 47 Hutton, M. 1980 Metal contamination of feral pigeons *Columba livia* from the London area: Part 2—Biological effects of lead exposure. *Environ. Pollut. (Series A)* **22**, 281-293.
- 48 Hutton, M. and Goodman, G. T. 1980 Metal contamination of feral pigeons *Columba livia* from the London area: Part 1—Tissue accumulation of lead, cadmium and zinc. *Environ. Pollut.* **22**, 207-217.
- 49 Ireland, M. P. and Wootton, R. J. 1977 Distribution of lead, zinc, copper and manganese in the marine gastropods *Thais lapillus* and *Littorina littorea*, around the coasts of Wales. *Environ. Pollut.* **12**, 27-41.
- 50 Jarvis, S. C. and Jones, L. H. P. 1980 The contents and sorption of cadmium in some agricultural soils of England and Wales. *J. Soil Sci.* **31**, 469-479.
- 51 Jarvis, S. C., Jones, L. H. P. and Hopper, M. J. 1976 Cadmium uptake from solution by plants and its transport from roots to shoots. *Plants and Soils* **44**, 179-191.
- 52 John, M. K. and Van Laerhoven, C. J. 1976 Differential effects of cadmium on lettuce varieties. *Environ. Pollut.* **10**, 163-173.
- 53 Johnson, M. S. and Eaton, J. W. 1980 Environmental contamination through residual trace metal dispersal from a derelict lead-zinc mine. *J. Environ. Quality* **9**, 175-179.
- 54 Johnson, M. S., Roberts, R. D., Hutton, M. and Inskip, M. J. 1978 Distribution of lead, zinc and cadmium in small mammals from polluted environments. *Oikos* **30**, 153-159.
- 55 Jung, J., Isermann, K. and Henjes, G. 1979 Einfluss von cadmiumhaltigen lünger-phosphaten auf die cadmiumanreicherung von culturböden und nutzpflanzen. *Landwirtsch. Forsch.* **32**, 262-274.
- 56 Kautz, K., Hirsch, H. and Singh Deo, R. 1975 Chemismus und Morphologie von Kraftwerksstäuben. VGB Kraftwerkstechnik **55**, 180-187.
- 57 Kjellström, T. 1980 Epidemiological aspects of the dose-response relationship of cadmium-induced renal damage. In: *Second International Cadmium Conference*, Cannes, February 1979. Metal Bulletin Ltd, 118-122.
- 58 Kjellström, T., Lind, B., Linnman, L. and Elinder, C-G. 1975 Variation of cadmium concentration in Swedish wheat and barley. *Arch. Environ. Health* **30**, 321-328.
- 59 Klein, D. H. and Russel, P. 1973 Heavy metals: fallout around a power plant. *Environ. Sci. Technol.* **7**, 357-358.

- 60 Kowal, N. E., Johnson, D. E., Kraemer, D. F. and Pahren, H. R. 1979 Normal levels of cadmium in diet, urine, blood, and tissues of inhabitants of the United States. *J. Toxicol. Environ. Health* **5**, 995-1014.
- 61 Kretzschmar, J. G., Delespaul, I. and De Rijck, T. H. 1980 Heavy metal levels in Belgium: a five-year survey. *Sci. Total Environ.* **14**, 85-97.
- 62 Lagerwerff, J. V. and Specht, A. D. 1970 Contamination of roadside soil and vegetation with cadmium, nickel, lead and zinc. *Environ. Sci. Technol.* **4**, 583-585.
- 63 Lauwerys, R. R. 1981 *Complement to criteria (dose/effect relationships) for cadmium*. To be published.
- 64 Lepow, M. L., Bruckman, L., Rubino, R. A., Markowitz, S., Gillette, M. and Kaphis, J. 1974 *Environ. Health Perspect.* Experimental Issue No. 7, 99-102.
- 65 Lindsay, D. G. 1979 Possible health hazards from the presence of persistent chemical residues in sewage sludge. In: *Utilisation of sewage sludge on land*, Oxford, 10-13 April 1978, Water Research Centre, Stevenage, Herts, 238-255.
- 66 Lindsay, D. G. 1980 Evaluation of the impact of inorganic soil pollutants on consumers. In: *Inorganic Pollution and Agriculture*, HMSO, London, 1-10.
- 67 Lyon, W. S. 1977 *Trace Element Measurements at the Allen Coal-Fired Steam Plant*. CRC Press, Cleveland, Ohio.
- 68 McLellan, J. S., Flanagan, P. R., Chamberlain, M. J. and Valberg, L. S. 1978 Measurements of dietary cadmium absorption in humans. *J. Toxicol. Environ. Health* **4**, 131-137.
- 69 Mack, D. and Schmid, R. 1979 Cadmium in nahrungspflanzen von kontaminierten standorten. *Dtsch. Lebensm.-Rdsch.* **75**, 309-311.
- 70 Martin, M. H., Coughtrey, P. J., Shales, S. W. and Little, P. 1980 Aspects of airborne cadmium contamination of soils and natural vegetation. In: *Inorganic Pollution and Agriculture*, HMSO, London, 56-69.
- 71 Miljøministeriet 1980 *Cadmiumforurening*. En redegørelse om anvendelse, forekomst og skadevirkninger af cadmium i Danmark. Stongard Jensen, København.
- 72 Minderhoud, A. and Boogerd, J. P. 1975 Determination of cadmium in air particulate matter by x-ray fluorescence. In: *Problems of the Contamination of Man and his Environment by Mercury and Cadmium*. Commission of the European Communities, Luxembourg, 203-209.
- 73 Ministry of Health, den Haag 1980 *Surveillance Programme Man and Nutrition*. State Supervisory Public Health Service.
- 74 Mouw, D. R., Kalitis, K., Anver, M., Schwartz, J., Constan, A., Hartung, R., Cohen, B. and Ringler, D. 1975 Lead: Possible toxicity in urban versus rural rats. *Arch. Environ. Health* **30**, 276-280.
- 75 Mulla, D. J., Page, A. L. and Ganje, T. J. 1980 Cadmium accumulation and bioavailability in soils from long-term phosphorus fertilization. *J. Environ. Qual.* **9**, 408-412.
- 76 Muller, J. and Beilke, S. 1977 Wet removal of heavy metals from the atmosphere. In: *International Conference on Heavy Metals in the Environment*, Toronto, Canada, October 27-31 1975, 987-999.
- 77 Muskett, C. J., Roberts, L. H. and Page, B. J. 1979 Cadmium and lead pollution from secondary metal refinery operations. *Sci. Total Environ.* **11**, 73-87.
- 78 Navarre, J.-L., Ronneau, C. and Priest, D. 1980 Deposition of heavy elements on Belgian agricultural soils. *Water, Air, Soil, Pollut.* **14**, 207-213.

- 79 Nriagu, J. O. 1979 Global inventory of natural and anthropogenic emissions of trace metals to the atmosphere. *Nature* **279**, 409-411.
- 80 OECD 1973 *Impact of fertilizers and agricultural waste products on the quality of waters*. Organisation for Economic Co-operation and Development Environment Directorate, Paris.
- 81 Official Journal of the European Communities 1979 *Coal policy: medium and long-term perspectives for coal in the Community*. No. C161/3. Commission of the European Communities, Brussels.
- 82 Oldham, R. G. and Wetherold, R. G. 1977 *Assessment, selection and development of procedures for determining the environmental acceptability of synthetic fuel plants*. National Technical Information Service. Report No. FE 1795-3.
- 83 Peterson, P. J. and Alloway, B. J. 1980 Cadmium in soils and vegetation. In: *The Chemistry, Biochemistry and Biology of Cadmium*, M. Webb (Ed.), Elsevier/North-Holland Biomedical Press, Amsterdam, 45-92.
- 84 Playford, K. and Pattenden, N. J. 1979 *Multi-element airborne dust measurements for Walsall Metropolitan Borough Report for year December 1977 to November 1978*. AERE Harwell Report G-1484. Harwell, Oxon.
- 85 Poelstra, P., Frissel, M. J. and El-Bassam, N. 1979 Transport and accumulation of Cd ions in soils and plants. *Z. Pflanzenernaehr. Bodenkd.* **142**, 848-864.
- 86 Provinciale Waterstaat van Noord-Holland 1975 *Onderzoek Regenwater in Noord-Holland, July-Dec. 1974*, Haarlem, The Netherlands.
- 87 Rahn, K. A. 1976 *The Chemical Composition of the Atmospheric Aerosol*. Technical Report, Graduate School of Oceanography, University of Rhode Island, Kingston, U.S.A.
- 88 Rauhut, A. 1978 *Survey of industrial emission of cadmium in the European Economic Community*. ENV/223/74/E. Commission of the European Communities, Brussels.
- 89 Roberts, R. D. and Johnson, M. S. 1978 Dispersal of heavy metals from abandoned mine workings and their transference through terrestrial food chains. *Environ. Pollut.* **16**, 293-310.
- 90 Roels, H. A., Lauwerys, R. R., Buchet, J-P. and Bernard, A. 1981 Environmental exposure to cadmium and renal function of aged women in three areas of Belgium. *Environ. Res.* (in press).
- 91 Röpenack, A. von 1978 Emission control in cadmium metal production. In: *First International Cadmium Conference, San Francisco*, January 1977, Metal Bulletin Ltd, 49-53.
- 92 Ruppert, H. 1975 Geochemical investigations on atmospheric precipitation in a medium-sized city (Göttingen, F.R.G.). *Water, Air, Soil, Pollut.* **4**, 447-460.
- 93 Scheltinga, H. M. J. 1979 Utilisation of sewage sludge on land in Holland. In: *Utilisation of Sewage Sludge on Land*, Oxford, 10-13 April 1978, 309-316. Water Research Centre, Stevenage, Herts.
- 94 Scott, D. W. 1979 *The fate of cadmium in municipal refuse incinerators*. Warren Spring Laboratory Report, CR 1710 (AP). Warren Springs, Stevenage, Herts.
- 95 Shipham Survey Committee 1980 *Soil Contamination at Shipham. Report on Studies Completed in the Village and Advice to Residents*, December 1980.
- 96 Simon, E. 1977 Cadmium tolerance in populations of *Agrostis tenuis* and *Festuca ovina*. *Nature* **265**, 328-330.

- 97 Solomon, R. L. and Hartford, J. W. 1976 Lead and cadmium in dusts and soils in a small urban community. *Environ. Sci. Technol.* **10**, 773-777.
- 98 Tjell, J. C., Hansen, J. Aa., Christensen, T. H. and Hovmand, M. F. 1981 Prediction of cadmium concentrations in Danish soils. In: *The Second European Symposium on Characterisation, Treatment and Use of Sewage Sludge*. Vienna, October 1980. P. L'Hermite and H. Ott (Eds) D. Reidel, Dordrecht, Holland, 652-664.
- 99 Thomas, B., Roughan, J. and Watters, E. D. 1972 Lead and cadmium content of some vegetable foodstuffs. *J. Sci. Food Agric.* **23**, 1493-1498.
- 100 Thormann, A. 1979 Utilisation of sewage sludge on land. The situation, problems and development trends in Germany. In: *Utilisation of Sewage Sludge on Land*, Oxford, 10-13 April 1978, 290-308. Water Research Centre, Stevenage, Herts.
- 101 Umweltbundesamt 1980 *Cadmium als Beispiel der Gesamtbelastung der Umwelt durch Schwermetalle*. Berlin, November 1980.
- 102 Van Enk, R. H. 1979 *The Pathway of Cadmium in the European Community*. EUR/6626/EN. Commission of the European Communities, Brussels.
- 103 Van Wambeke, L. 1979 *Measures taken by the European Communities in respect of cadmium between 1977 and 1979 and inventory of emissions into the environment*. ENV/322/79/FR. Commission of the European Communities, Brussels.
- 104 Van Wambeke, L. 1980 *Energy from municipal waste in the European Community*. CEC XII/252/80-EN.
- 105 Webber, M. D. 1979 *Phosphate Fertilizer and Sewage Sludge use on Agricultural Land—The Potential for Cadmium Uptake by Crops*. Environment Canada Technology Development Report EPS 4-WP-79-2. Environment Canada, Ottawa.
- 106 Webber, L. R. and Beauchamp, E. G. 1979 Cadmium concentration and distribution in corn (*Zea mays* L.) grown on a calcareous soil for three years after three annual sludge applications. *J. Environ. Sci. Health* **B14**, 459-474.
- 107 Welsh Office 1975 *Report of a collaborative study on certain elements in air, soil, plants, animals and humans in the Swansea/Neath/Port Talbot area, together with a report on a moss bag study of atmospheric pollution across South Wales*. The Welsh Office, Cardiff.
- 108 Williams, C. H. and David, D. J. 1973 The effect of superphosphate on the cadmium content of soil and plants. *Aust. J. Soil Res.* **11**, 43-56.
- 109 Williams, C. H. and David, D. J. 1976 The accumulation in soil of cadmium residues from phosphate fertilizers and their effect on the cadmium content of plants. *Soil Sci.* **121**, 86-93.
- 110 Williams, D. E., Vlamis, J., Pukite, A. H. and Corey, J. E. 1980 Trace element accumulation, movement, and distribution in the soil profile from massive applications of sewage sludge. *Soil Sci.* **129**, 119-132.
- 111 Williamson, P. 1980 Variables affecting body burdens of lead, zinc and cadmium in a roadside population of the snail *Cepaea hortensis* Müller. *Oecologia* (Berl.) **44**, 213-220.
- 112 Yost, K. J. 1979 Some aspects of cadmium flow in the U.S. *Environ. Health Perspect.* **28**, 5-16.
- 113 Yost, K. J. and Miles, L. J. 1979 Environmental health assessment for cadmium: a systems approach. *J. Environ. Sci. Health* **A14**, 285-311.

# APPENDIX 1 Primary zinc production in the EC

| Country             | Plant location         | Process | Zinc production ( $10^3$ t) |                  |
|---------------------|------------------------|---------|-----------------------------|------------------|
|                     |                        |         | Annual capacity             | 1979             |
| Italy               | Porto Marghera, Venice | ER      | 45                          | 160 <sup>a</sup> |
|                     | Ponte Nossà, Bergamo   | ER      | 35                          |                  |
|                     | Cagliari, Sardinia     | ER      | 15                          |                  |
|                     | Crotone, Calabria      | ER      | 90                          |                  |
|                     | Porto Vesme, Sardinia  | ISF     | 50                          | 43 <sup>a</sup>  |
| The Netherlands     | Budel                  | ER      | 160                         | 153.9            |
| U.K.                | Avonmouth              | ISF     | 100                         | 77.4             |
| F.R.G. <sup>b</sup> | Datteln                | ER      | 140                         | 199.3            |
|                     | Nordenham              | ER      | 110                         |                  |
|                     | Duisburg               | ISF     | 80                          | 75               |
|                     | Harlingerode           | VR      | 100                         | 81.2             |
| Belgium             | Balen                  | ER      | 170                         | 252.6            |
|                     | Overpelt               | ER      | 120                         |                  |
|                     | Ehein                  | ER      | 70                          |                  |
| France              | Auby-les-Douai         | ER      | 110                         | 163              |
|                     | Viviez, Aveyron        | ER      | 94                          |                  |
|                     | Noyelles Godault       | ISF     | 105                         | 86               |

<sup>a</sup> Estimated values, taking into account relative production capacities of each plant and actual production in 1979.

<sup>b</sup> An electrothermal plant at Duisberg with a zinc production capacity of  $10 \times 10^3$  t Zn y<sup>-1</sup> has not been considered in this analysis.

ER—Electrolytic refinery.

ISF—Imperial Smelting Furnace smelter.

VR—Vertical retort smelter.

Sources: Hiscock (1978); World Metal Statistics (1980); and personal communications from company employees.

## APPENDIX 2 Cadmium in U.K. steelmaking operations

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### 1 Introduction

To understand the behaviour of cadmium through the steel industry it is necessary to know the quantity of cadmium associated with the raw materials, products and wastes involved at each stage. This, in turn, requires a measure of cadmium concentrations and a knowledge of the quantities of all the materials and wastes concerned. It is then possible to construct a mass balance for the metal.

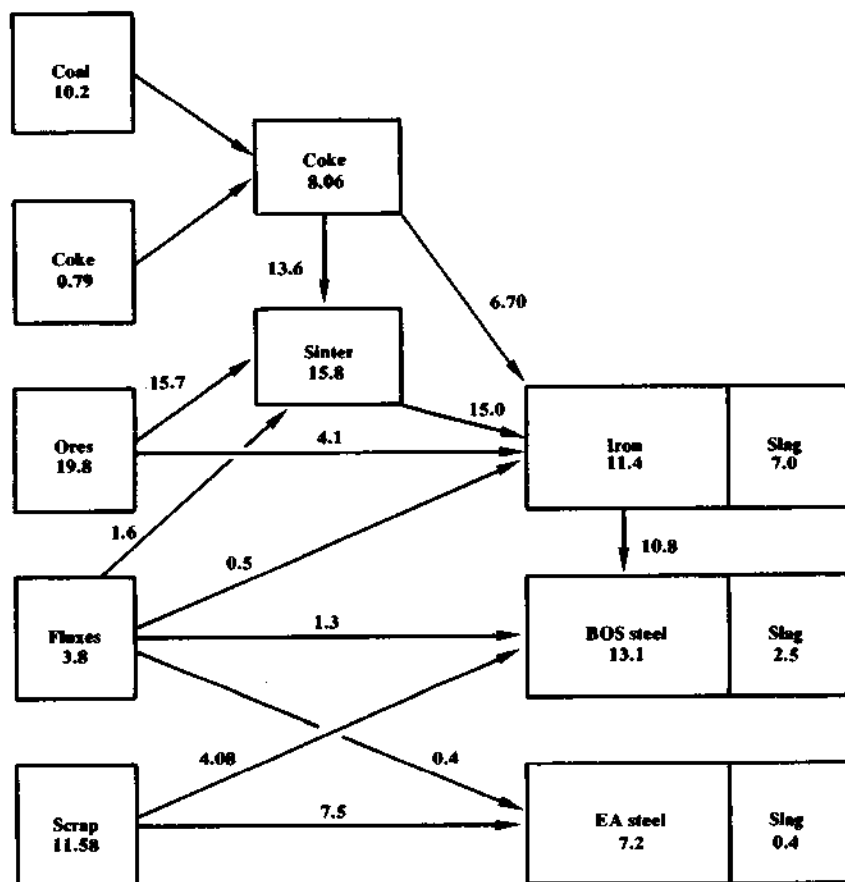
The information presented here has come primarily from the collection and analysis of samples of materials taken from a large number of U.K. plants, particularly in the steelmaking area. Samples were collected on a random basis, and no attempt was made to obtain composite samples or samples representative of short-term variations in cadmium concentrations (as is known can occur, particularly with steelmaking fume).

Despite the relatively large number of samples analysed altogether, the number associated with a particular source was generally quite small, especially in statistical terms. Nevertheless, it is felt that a reasonable compromise has been made between expediency and accuracy. The

**Table 1** Concentrations of cadmium in various materials associated with U.K. steelmaking

| Material                                    | Number of sources sampled | Mean cadmium concentration ( $\mu\text{g g}^{-1}$ ) |
|---------------------------------------------|---------------------------|-----------------------------------------------------|
| Iron ore                                    | several                   | 0.4                                                 |
| Coal                                        | —                         | 1                                                   |
| Limestone                                   | —                         | 0.035*                                              |
| Coke oven effluent                          | 1                         | 0.02 ( $\text{mg l}^{-1}$ )                         |
| Dust to atmosphere from sinter plants       | 3                         | 50                                                  |
| Solids from blast furnace gas washing water | 5                         | 42                                                  |
| Basic oxygen steelmaking fume               | 8                         | 31                                                  |
| Electric arc steelmaking fume               | 5                         | 317                                                 |
| Basic oxygen steelmaking slag               | 4                         | 5                                                   |
| Electric arc steelmaking slag               | 5                         | 5                                                   |
| Steel                                       | 5                         | <0.05                                               |

\* Previously published data.



**Figure 1** Flow diagram for the major raw materials and products in the U.K. steel industry in 1978

Note: Figures are in millions of tonnes; those in boxes represent total usage or production, while those on arrows indicate the consumption in specific processes.

samples involved have been solids, slurries or effluents, and after filtering and drying as necessary, were analysed for cadmium, by atomic absorption spectrophotometry.

Tonnages of materials consumed and produced have mostly been taken from published statistics of the U.K. steel industry; figures for the year 1978 have been used, and so all the calculated flows for cadmium relate to this year.

## 2 Results

Table 1 summarizes concentration data for cadmium arising from this study, while Figure 1 gives the tonnages of materials used in the U.K. steel industry in 1978. Combining these two sets of information it is possible to construct Figure 2—a flow diagram for cadmium and from this the summary data in Table 2 are calculated.

## 3 Discussion

Total atmospheric emissions are similar to those from the cadmium/zinc smelting industry. Discharges of particulates containing cadmium from the latter are reflected in elevated cadmium concentrations in soils near to the single U.K. plant involved (at Avonmouth). The 4.5 tonnes from the steel industry, however, come from widespread sources across the country, and are associated with much larger quantities of fine particulate

**Table 2 Summary of potential cadmium discharges to the environment from the U.K. steel industry for 1978 (tonnes)**

| Process area             | Atmospheric emissions | Liquid effluents | Solid wastes | Slags       |
|--------------------------|-----------------------|------------------|--------------|-------------|
| Cokemaking               | 0.15                  | 0.20             | *            | —           |
| Sinter production        | 1.2                   | —                | —            | —           |
| Ironmaking               | —                     | *                | 7.6          | *           |
| Basic oxygen steelmaking | 0.25                  | *                | 4.7          | 14.5        |
| Electric arc steelmaking | 2.9                   | *                | 26.3         | 2.1         |
| <b>Total</b>             | <b>4.5</b>            | <b>0.20</b>      | <b>38.6</b>  | <b>16.6</b> |

Grand total = 59.9 tonnes

\* indicates that no data are available but quantities are likely to be very small.

— indicates that no such discharges occur from this process area.

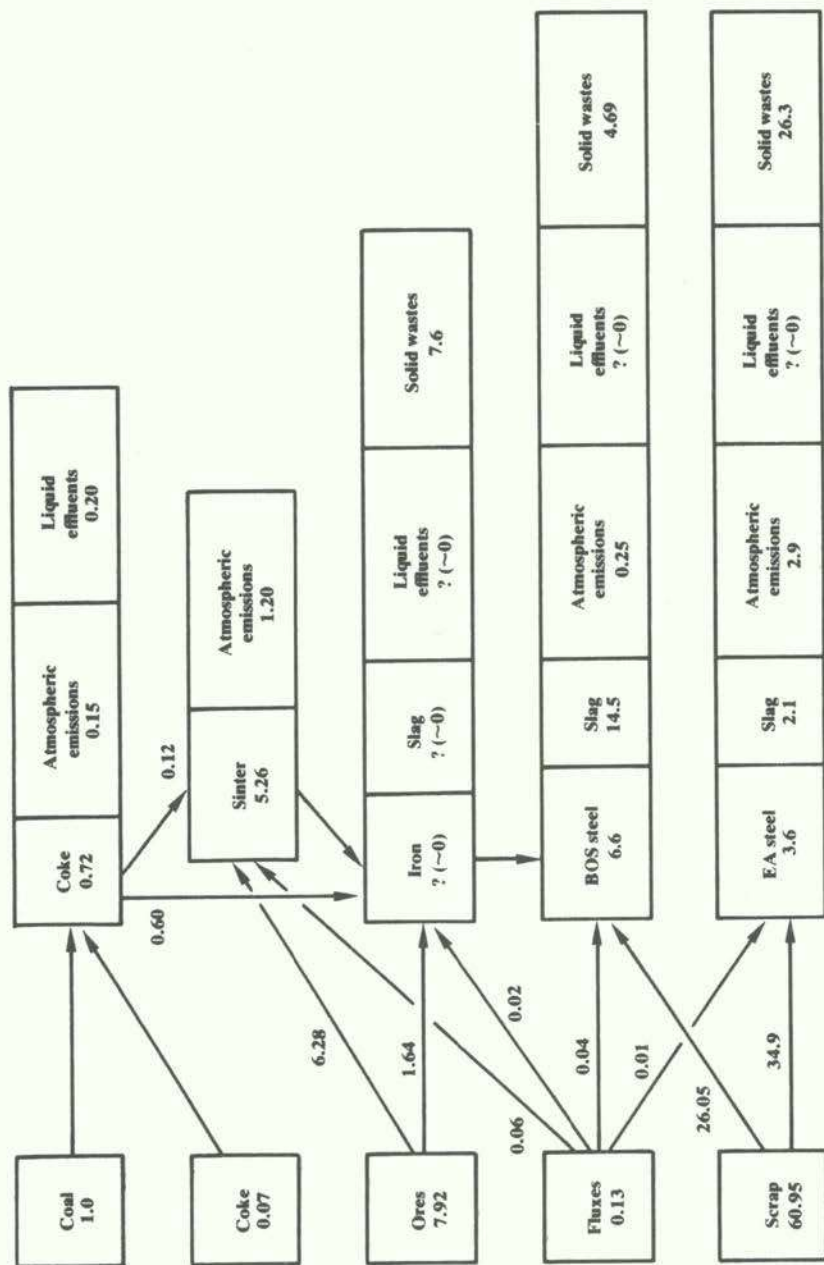


Figure 2 Flow diagram for cadmium in the U.K. steel industry  
Note: Figures are in tonnes, based on 1978 production data.

material. Consequently, deposition will take place over a very wide area and will be of particulates containing low concentrations of cadmium relative to the cadmium-zinc smelter emissions. Thus no pollution of soils is likely from cadmium in dustfall from the steel industry.

The atmospheric dispersion of particulates discharged from the industry will depend very much on local circumstances and so it is not possible to predict whether an increased level of cadmium in air will result from such discharges. A study of airborne cadmium in the Chicago area of the U.S.A. has failed to detect any elevated concentrations, despite numerous local steel scrap smelters. On this basis, it seems unlikely that the U.K. steel industry makes a significant contribution to atmospheric cadmium levels.

Few data are available for cadmium discharges in liquid effluents. The metal will be present in such effluents as either an insoluble suspension or in very dilute solution. Provided that there is dispersion of the effluent in the receiving water, then it seems very unlikely that any environmental problems should occur.

Solid wastes from iron and steelmaking processes account for about 65 per cent of the total discharges from the industry. While many of these wastes are not suitable for recycling through production processes because of their content of tramp elements (zinc especially), they are usually stockpiled or dumped within the industry's sites. In this way they are unlikely to become a potential environmental problem.

#### 4 Conclusions

In 1978, approximately 70 tonnes of cadmium were introduced to the U.K. steel industry's production plants in its primary and recycled raw materials. Of this, about 10.2 tonnes (14.6 per cent) was translocated into the steel product, the remainder occurring in atmospheric emissions (4.5 tonnes, 6.4 per cent), liquid effluents (0.2 tonnes, 0.3 per cent), solid wastes (38.6 tonnes, 55.1 per cent) and slags (16.6 tonnes, 23.7 per cent).

There are unlikely to be any significant air or water pollution problems involving cadmium as a result of U.K. steelmaking operations. However, slurries and dusts from iron and steel plant gas washing systems can contain sufficient cadmium to warrant care in their disposal. There is no evidence at present that environmental pollution is occurring as a result of the dumping or stockpiling of these materials.

### APPENDIX 3 Estimated discharges of cadmium from the EC iron and steel industries in 1979

| Country         | Process             | Production<br>1979<br>( $\times 10^6$ t) | Cadmium in discharge medium<br>(tonnes) |                 |       |
|-----------------|---------------------|------------------------------------------|-----------------------------------------|-----------------|-------|
|                 |                     |                                          | Atmospheric<br>emissions                | Solid<br>wastes | Slags |
| Italy           | Sinter <sup>a</sup> | 14.8                                     | 1.2                                     | —               | —     |
|                 | Pig iron            | 11.3                                     | —                                       | 7.6             | —     |
|                 | BOS                 | 10.2                                     | 0.2                                     | 3.7             | 11.3  |
|                 | EAS                 | 12.9                                     | 5.2                                     | 47.0            | 3.7   |
|                 | OH                  | 1.1                                      | 1.1                                     | ND              | ND    |
|                 |                     |                                          | 7.7                                     | 58.3            | 15.0  |
| Luxembourg      | Sinter              | 7.4                                      | 0.6                                     | —               | —     |
|                 | Pig iron            | 3.8                                      | —                                       | 2.6             | —     |
|                 | BOS                 | 5.0                                      | 0.1                                     | 1.8             | 5.5   |
|                 |                     |                                          | 0.7                                     | 4.4             | 5.5   |
| The Netherlands | Sinter              | 2.9                                      | 0.85 <sup>b</sup>                       | —               | —     |
|                 | Pig iron            | 4.8                                      | —                                       | 3.2             | —     |
|                 | BOS                 | 5.5                                      | 0.05 <sup>b</sup>                       | 2.0             | 6.1   |
|                 |                     |                                          | 0.9                                     | 5.2             | 6.1   |
| U.K.            | Sinter              | 17.6                                     | 1.4                                     | —               | —     |
|                 | Pig iron            | 12.9                                     | —                                       | 8.6             | —     |
|                 | BOS                 | 12.9                                     | 0.3                                     | 4.6             | 14.3  |
|                 | EAS                 | 7.4                                      | 3.0                                     | 27.0            | 2.1   |
|                 | OH                  | 1.2                                      | 1.2                                     | ND              | ND    |
|                 |                     |                                          | 5.9                                     | 40.2            | 16.4  |
| F.R.G.          | Sinter              | 36.0                                     | 2.9                                     | —               | —     |
|                 | Pig iron            | 35.2                                     | —                                       | 23.6            | —     |
|                 | BOS                 | 35.0                                     | 0.7                                     | 12.6            | 39.0  |
|                 | EAS                 | 6.4                                      | 2.5                                     | 23.3            | 1.9   |
|                 | OH                  | 4.6                                      | 4.6                                     | ND              | ND    |
|                 |                     |                                          | 10.7                                    | 59.5            | 40.9  |

**APPENDIX 3** (continued)

| Country  | Process  | Production<br>1979<br>( $\times 10^6$ t) | Cadmium in discharge medium<br>(tonnes) |                   |       |
|----------|----------|------------------------------------------|-----------------------------------------|-------------------|-------|
|          |          |                                          | Atmospheric<br>emissions                | Solid<br>wastes   | Slags |
| Belgium  | Sinter   | 11.4 <sup>c</sup>                        | 0.9                                     | —                 | —     |
|          | Pig iron | 10.8                                     | —                                       | 7.2               | —     |
|          | BOS      | 12.9                                     | 0.3                                     | 4.6               | 14.3  |
|          | EAS      | 0.6                                      | 0.2                                     | 2.2               | 0.2   |
|          |          |                                          | 1.4                                     | 14.0              | 14.5  |
| France   | Sinter   | 33.7                                     | 2.7                                     | —                 | —     |
|          | Pig iron | 19.4                                     | —                                       | 13.0              | —     |
|          | BOS      | 18.6                                     | 0.4                                     | 6.7               | 20.7  |
|          | EAS      | 3.6                                      | 2.8 <sup>d</sup>                        | 26.3 <sup>d</sup> | 1.0   |
|          | OH       | 0.4                                      | 0.4                                     | ND                | ND    |
|          | BB       | 0.8 <sup>e</sup>                         | ND                                      | ND                | ND    |
|          |          |                                          | 6.3                                     | 46.0              | 21.7  |
| Denmark  | EAS      | 0.4                                      | 0.2                                     | 1.5               | 0.1   |
|          | OH       | 0.3                                      | 0.3                                     | ND                | ND    |
|          |          |                                          | 0.5                                     | 1.5               | 0.1   |
| EC Total |          |                                          | 34                                      | 229               | 120   |

1979 Production data from International Iron and Steel Institute.

<sup>a</sup> 1978 sinter production.

<sup>b</sup> Atmospheric emission estimates supplied by C. J-P. Steiner.

<sup>c</sup> 1977 sinter production.

<sup>d</sup> Emission factors for atmospheric emissions and solid wastes from EAS doubled for France.

<sup>e</sup> No discharge estimates made for Basic Bessemer steelmaking.

BOS—Basic Oxygen Steelmaking.

EAS—Electric Arc Steelmaking.

OH—Open Hearth steelmaking.

BB—Basic Bessemer steelmaking.

ND—Not determined.

# **APPENDIX 4 Predicted discharges of cadmium from the EC iron and steel industries in 1990 and 2000**

| Country         | Process  | Predicted production<br>( $\times 10^6$ t)<br>1990/2000 | Cadmium in discharge medium (tonnes) |              |           |
|-----------------|----------|---------------------------------------------------------|--------------------------------------|--------------|-----------|
|                 |          |                                                         | Atmospheric emissions                | Solid wastes | Slags     |
| Italy           | Sinter   | 18.4/22.6                                               | 1.5/1.8                              | —            | —         |
|                 | Pig iron | 14.0/17.1                                               | —                                    | 9.4/11.5     | —         |
|                 | BOS      | 12.6/15.3                                               | 0.2/0.3                              | 4.5/5.5      | 14.0/17.0 |
|                 | EAS      | 17.4/21.2                                               | 7.0/8.5                              | 63.5/77.0    | 5.1/6.2   |
|                 |          |                                                         | 8.7/10.6                             | 77.4/94.0    | 19.1/23.2 |
| Luxembourg      | Sinter   | 9.2/11.2                                                | 0.7/0.9                              | —            | —         |
|                 | Pig iron | 4.7/5.8                                                 | —                                    | 3.1/3.9      | —         |
|                 | BOS      | 6.2/7.5                                                 | 0.1/0.2                              | 2.2/2.7      | 6.9/8.3   |
|                 |          |                                                         | 0.8/1.1                              | 5.3/6.6      | 6.9/8.3   |
| The Netherlands | Sinter   | 3.6/4.4                                                 | 1.1/1.3                              | —            | —         |
|                 | Pig iron | 6.0/7.3                                                 | —                                    | 4.0/4.9      | —         |
|                 | BOS      | 6.8/8.3                                                 | 0.06/0.08                            | 2.5/3.0      | 7.6/9.2   |
|                 |          |                                                         | 1.2/1.4                              | 6.5/7.9      | 7.6/9.2   |
| U.K.            | Sinter   | 21.9/26.7                                               | 1.8/2.1                              | —            | —         |
|                 | Pig iron | 16.0/19.6                                               | —                                    | 10.7/13.1    | —         |
|                 | BOS      | 16.0/19.6                                               | 0.3/0.4                              | 5.8/7.0      | 17.8/21.7 |
|                 | EAS      | 10.6/13.0                                               | 4.2/5.2                              | 39.0/47.0    | 3.1/3.8   |
|                 |          |                                                         | 6.3/7.7                              | 55.5/67.1    | 20.9/25.5 |
| F.R.G.          | Sinter   | 44.8/54.7                                               | 3.6/4.4                              | —            | —         |
|                 | Pig iron | 43.7/53.4                                               | —                                    | 29.0/35.8    | —         |
|                 | BOS      | 43.3/52.7                                               | 0.9/1.0                              | 15.6/19.0    | 48.0/58.5 |
|                 | EAS      | 13.7/16.7                                               | 5.5/6.7                              | 50.0/61.0    | 4.0/4.8   |
|                 |          |                                                         | 10.0/12.1                            | 94.6/115.8   | 52.0/63.3 |
| Belgium         | Sinter   | 14.2/17.3                                               | 1.1/1.4                              | —            | —         |
|                 | Pig iron | 13.4/16.4                                               | —                                    | 9.0/11.0     | —         |
|                 | BOS      | 16.0/19.5                                               | 0.3/0.4                              | 5.8/7.0      | 17.8/21.7 |
|                 | EAS      | 0.7/0.9                                                 | 0.3/0.4                              | 2.6/3.3      | 0.2/0.3   |
|                 |          |                                                         | 1.7/2.2                              | 17.4/21.3    | 18.0/22   |

**APPENDIX 4** *(continued)*

| Country            | Process  | Predicted<br>production<br>( $\times 10^6$ t)<br>1990/2000 | Cadmium in discharge medium (tonnes) |                 |             |
|--------------------|----------|------------------------------------------------------------|--------------------------------------|-----------------|-------------|
|                    |          |                                                            | Atmospheric<br>emissions             | Solid<br>wastes | Slags       |
| France             | Sinter   | 41.9/51.2                                                  | 3.4/4.1                              | —               | —           |
|                    | Pig iron | 24.1/29.4                                                  | —                                    | 16.1/19.7       | —           |
|                    | BOS      | 23.2/28.4                                                  | 0.5/0.6                              | 8.4/10.2        | 25.8/31.5   |
|                    | EAS      | 5.8/7.1                                                    | 4.6/5.7                              | 42.3/51.8       | 1.7/2.1     |
|                    |          |                                                            | 8.5/10.4                             | 66.8/81.7       | 27.5/33.6   |
| Denmark            | EAS      | 0.9/1.0                                                    | 0.4/0.4                              | 3.3/3.7         | 0.3/0.3     |
| EC Total 1990/2000 |          |                                                            | 37.6/45.9                            | 326.8/398       | 152.3/185.4 |

BOS—Basic Oxygen Steelmaking; EAS—Electric Arc Steelmaking.

**APPENDIX 5 Pattern of rock phosphate importation, fertilizer consumption and estimated cadmium discharges to agricultural land in the EC for 1979**

| Country                | Fertilizer use for 1979/80 ( $\times 10^3$ t $P_2O_5$ ) | Rock phosphate imports 1979 ( $\times 10^3$ t) | Assumed fertilizer use according to country ( $\times 10^3$ t $P_2O_5$ ) | Estimated Cd discharges to land (t) |
|------------------------|---------------------------------------------------------|------------------------------------------------|--------------------------------------------------------------------------|-------------------------------------|
| Italy                  | 890                                                     | U.S.A. 340                                     | 174                                                                      | 6                                   |
|                        |                                                         | Morocco                                        |                                                                          |                                     |
|                        |                                                         | Tunisia } 891                                  | 454                                                                      | 27                                  |
|                        |                                                         | Algeria }                                      |                                                                          |                                     |
|                        |                                                         | Togo 106                                       | 53                                                                       | 8.6                                 |
|                        |                                                         | Israel } 409                                   | 209                                                                      | 7.3                                 |
|                        |                                                         | Jordan }                                       |                                                                          |                                     |
|                        |                                                         |                                                |                                                                          | 48.9                                |
| The Netherlands        | 85                                                      | U.S.A. 608                                     | 21                                                                       | 0.75                                |
|                        |                                                         | Morocco 935                                    | 33                                                                       | 2.0                                 |
|                        |                                                         | Togo 681                                       | 24                                                                       | 3.8                                 |
|                        |                                                         | Israel 200                                     | 7                                                                        | 0.25                                |
|                        |                                                         |                                                |                                                                          | 6.8                                 |
| U.K.                   | 449                                                     | U.S.A. 411                                     | 108                                                                      | 3.8                                 |
|                        |                                                         | Morocco } 905                                  | 233                                                                      | 14                                  |
|                        |                                                         | Tunisia }                                      |                                                                          |                                     |
|                        |                                                         | Senegal 415                                    | 108                                                                      | 27.6                                |
|                        |                                                         |                                                |                                                                          | 45.4                                |
| F.R.G.                 | 914                                                     | U.S.A. 1,480                                   | 466                                                                      | 16.3                                |
|                        |                                                         | U.S.S.R.* 581                                  | 183                                                                      | 0.15                                |
|                        |                                                         | Morocco } 422                                  | 132                                                                      | 7.7                                 |
|                        |                                                         | Tunisia }                                      |                                                                          |                                     |
|                        |                                                         | Senegal 174                                    | 54.8                                                                     | 14                                  |
|                        |                                                         | Togo 86                                        | 27                                                                       | 4.3                                 |
|                        |                                                         | Israel 165                                     | 52                                                                       | 1.8                                 |
|                        |                                                         |                                                |                                                                          | 44.2                                |
| Belgium/<br>Luxembourg | 118                                                     | U.S.A. 260                                     | 14                                                                       | 0.5                                 |
|                        |                                                         | Morocco } 1,533                                | 87                                                                       | 5                                   |
|                        |                                                         | Tunisia }                                      |                                                                          |                                     |
|                        |                                                         | Togo 233                                       | 13                                                                       | 2.1                                 |
|                        |                                                         | Israel 59                                      | 3                                                                        | 0.1                                 |
|                        |                                                         |                                                |                                                                          | 7.7                                 |
| France                 | 2,018                                                   | U.S.A. 983                                     | 379                                                                      | 13.3                                |
|                        |                                                         | Morocco                                        |                                                                          |                                     |
|                        |                                                         | Tunisia } 2,976                                | 1,150                                                                    | 69                                  |
|                        |                                                         | Algeria }                                      |                                                                          |                                     |
|                        |                                                         | Senegal 141                                    | 54.5                                                                     | 14                                  |
|                        |                                                         | Togo 687                                       | 262                                                                      | 42                                  |
|                        |                                                         | Israel } 435                                   | 167                                                                      | 5.9                                 |
|                        |                                                         | Jordan }                                       |                                                                          |                                     |
|                        |                                                         |                                                |                                                                          | 144                                 |

**APPENDIX 5** (continued)

| Country                | Fertilizer<br>use for<br>1979/80<br>( $\times 10^3$ t<br>$P_2O_5$ ) | Rock phosphate<br>imports 1979<br>( $\times 10^3$ t) |     | Assumed<br>fertilizer<br>use according<br>to country<br>( $\times 10^3$ t $P_2O_5$ ) | Estimated<br>Cd<br>discharges<br>to land<br>(t) |     |
|------------------------|---------------------------------------------------------------------|------------------------------------------------------|-----|--------------------------------------------------------------------------------------|-------------------------------------------------|-----|
| Denmark                | 133                                                                 | U.S.A.                                               | 87  | 41                                                                                   | 1.4                                             | 6.9 |
|                        |                                                                     | Morocco                                              | 193 | 92                                                                                   | 5.5                                             |     |
| Republic of<br>Ireland | 156                                                                 | U.S.A.                                               | 31  | 41                                                                                   | 1.4                                             | 8.3 |
|                        |                                                                     | Morocco                                              | 86  | 115                                                                                  | 6.9                                             |     |
| EC Total               |                                                                     |                                                      |     |                                                                                      | 312                                             |     |

Rock phosphate import data from *Phosphorus & Potassium* No. 106, March-April 1980.

\* Imports of rock phosphate from the U.S.S.R. assumed to be 20 per cent of total.

## APPENDIX 6 Cadmium contamination at Shipham—a summary of published information

Shipham, a small village (population 1,092) located in Somerset, South West England, is the site of an abandoned zinc mine, last worked in the 19th century. The form of zinc mined at Shipham, calamine ( $\text{ZnCO}_3$ ), is unusual as most zinc elsewhere in the U.K. is present as sphalerite ( $\text{ZnS}$ ). The village has expanded in recent years and contains large numbers of spoil heaps from the mine workings.

A study of the village by Davies and Ginnever (1979) revealed that cadmium levels in the spoil heaps were very high, with a maximum of  $997 \mu\text{g g}^{-1}$  cadmium. The same study reported that the total area contaminated (defined as soil cadmium levels greater than  $10 \mu\text{g g}^{-1}$ ) was  $2 \text{ km}^2$ . Soil levels in gardens averaged  $69.5 \mu\text{g g}^{-1}$ , more than 30 times the control value. A much larger survey, conducted by the U.K. Department of the Environment (DOE) sampled soils from over 300 households. Levels in vegetable plots were highly variable, ranging from 2 to  $520 \mu\text{g g}^{-1}$ , with an average of  $87 \mu\text{g g}^{-1}$ .

Davies and Ginnever (1979) also measured cadmium levels in three vegetable types grown in Shipham gardens; their findings are summarized below.

| Garden | Soil cadmium<br>$\mu\text{g g}^{-1}$ dry<br>weight | Cadmium $\mu\text{g g}^{-1}$ fresh weight |                    |                     |
|--------|----------------------------------------------------|-------------------------------------------|--------------------|---------------------|
|        |                                                    | Potatoes                                  | Root<br>vegetables | Brussels<br>sprouts |
| 1      | 8.8                                                | 0.02                                      | 0.15               | 0.44                |
| 2      | 69.9                                               | 0.07                                      | 0.07               | 0.07                |
| 3      | 70                                                 | 0.08                                      | 0.097              | 0.04–0.09           |
| 4      | 80.8                                               | 0.15                                      | —                  | —                   |
| 5      | 70.2                                               | 0.057                                     | 0.05               | 0.03                |
| 6      | 88.8                                               | —                                         | 0.46               | 0.10                |
|        |                                                    | 0.08                                      | 0.17               | 0.14                |

It is of interest to note that the extent of cadmium contamination in vegetables does not always correspond to the soil level and that an elevated cadmium level in one vegetable species is not necessarily reflected by a similar value in another species. Thus, soil cadmium in Garden 1 was only  $8.8 \mu\text{g g}^{-1}$  yet cadmium levels in the Brussels sprouts

were the highest analysed. Further, Garden 6 produced root vegetables with the highest cadmium levels, yet values in Brussels sprouts were relatively low.

It has been estimated that on average 350 g vegetables are consumed per person daily in the U.K. If it is assumed that locally grown vegetables are consumed exclusively then potatoes alone would on average supply  $196 \mu\text{g w}^{-1}$ , root vegetables alone  $416 \mu\text{g w}^{-1}$  and Brussels sprouts alone  $343 \mu\text{g w}^{-1}$ . Note that these figures refer to the average values. If the maximum value for each vegetable is used then potatoes alone (from Garden 4) would supply  $367 \mu\text{g w}^{-1}$ , root vegetables (from Garden 6)  $1,127 \mu\text{g w}^{-1}$ , and Brussels sprouts (from Garden 1)  $1,078 \mu\text{g w}^{-1}$ . The WHO provisional tolerable intake for cadmium from all sources is  $400\text{--}500 \mu\text{g w}^{-1}$ .

The larger DOE survey also measured cadmium levels in locally grown vegetables. Concentrations averaged  $0.28 \mu\text{g g}^{-1}$  wet weight prepared for eating (range  $0.02\text{--}1.77 \mu\text{g g}^{-1}$ ) in 168 winter crops and  $0.52 \mu\text{g g}^{-1}$  (range  $0.01\text{--}3.56 \mu\text{g g}^{-1}$ ) in 215 summer crops.

Detailed dietary studies of Shiphams residents are being carried out by the DOE. In a smaller, independent survey, Carruthers and Smith (1979) reported that on average 50 per cent of vegetables consumed by Shiphams residents were home-grown. Additionally, in 4 of the 31 residents sampled, 100 per cent of vegetables eaten were home-grown.

These findings indicate that a proportion of the population in Shiphams regularly exceeds the WHO tolerable intake figures from the consumption of vegetables alone. The ingestion of dusts derived from the spoil heaps may also be a particularly important source of cadmium for children in the area. The concentrations of cadmium as well as lead in household dusts are being measured by the DOE.

Liver cadmium levels have been measured in 21 adults living in Shiphams by an *in vivo* neutron activation technique (Harvey *et al.* 1979). The mean value,  $11.0 \pm 2.0 \mu\text{g g}^{-1}$  (wet weight), was significantly higher than the  $2.2 \pm 2.0 \mu\text{g g}^{-1}$  found in 10 control subjects. Although these values are about five times higher than controls, they are much lower than those found in occupationally-exposed workers.

A small clinical and biochemical survey of 31 volunteers from Shiphams has recently been reported (Carruthers and Smith 1979). The major findings were elevated blood cadmium levels, increased prevalence of both hypertension and  $\beta_2$ -microglobulin excretion. These findings have, however, been questioned by Hughes and Stewart (1979). It was pointed

out that the survey's sample was not randomly selected but biased towards the anxious or sick. For this reason the reported prevalence of hypertension in Shipham residents was dismissed. Hughes and Stewart also pointed out that the extent of  $\beta_2$ -microglobulin excretion could not be related to the duration of residence in Shipham. Furthermore, it was noted that there was no relationship between blood cadmium levels and the extent of  $\beta_2$ -microglobulin excretion.

It is suggested that any conclusions regarding the health of Shipham residents must await the detailed epidemiological survey being carried out by the DOE.

## References

- Carruthers, M. and Smith, B. 1979 Evidence of cadmium toxicity in a population living in a zinc mining area. *Lancet*, 845-847.
- Davies, B. E. and Ginnever, R. C. 1979 Trace metal contamination of soils and vegetables in Shipham, Somerset. *J. Agric. Sci., Camb.* **93**, 753-756.
- Harvey, T. C., Chettle, D. R., Frelmin, J. H., Al Haddad, I. K. and Downey, S. P. M. 1979 Cadmium in Shipham. *Lancet*, 557.
- Hughes, E. G. and Stewart, M. 1979 Cadmium in Shipham. *Lancet*, 973-974.

## APPENDIX 7 Cadmium in soil-plant systems. Derivation of expressions for transfer coefficients and mean residence times

### Introduction

The uptake of cadmium by plants on a unit area basis is given by

$$F_{SP} = ST(\alpha C_S)\sigma/(\sigma + \theta\alpha) \quad (1)$$

The rate of loss of cadmium from soil as a result of plant uptake and runoff is given by

$$F_{SP} + F_{SO} = (ST + L)(\alpha C_S)\sigma/(\sigma + \theta\alpha) \quad (2)$$

where  $C_S$  is the cadmium concentration in soil particulates (e.g. in  $\mu\text{g g}^{-1}$ ). The surface density of cadmium in soil particulates,  $C'_S$ , (e.g. in  $\text{kg ha}^{-1}$ ) is given by the expression

$$C'_S = C_S\sigma d \quad (3)$$

The mean residence time for cadmium in the soil particulates fraction is defined as the ratio of the soil cadmium surface density and the rate of loss by all routes (uptake by plants and runoff)

$$G'_S = C'_S/(F_{SP} + F_{SO}) = \frac{C_S\sigma d}{(ST + L)(\alpha C_S)\sigma/(\sigma + \theta\alpha)}$$

or

$$G'_S = \frac{(\sigma + \theta\alpha)d}{\alpha(ST + L)} \quad (4)$$

If it is assumed that the ratio of the rate of transpiration of plants,  $T$ , and the rate of creation of plant material is a simple constant,  $\beta$  (expressed in  $\text{g y}^{-1} \text{ha}^{-1}$  per  $\text{g y}^{-1} \text{ha}^{-1}$ ), the plant cadmium concentration associated with the flux,  $F_{SP}$ , is given, using equation (1) above by

$$C_P = F_{SP}/(T/\beta) = \beta F_{SP}/T$$

i.e.

$$C_P = \frac{\beta ST(\alpha C_S)\sigma}{T(\sigma + \theta\alpha)}$$

However, the transfer coefficient  $P_{SP}$  is defined as  $C_P/C_S$ , therefore

$$P_{SP} = \frac{\beta S\alpha\sigma}{(\sigma + \theta\alpha)} \quad (5)$$

For a given input rate of cadmium to soil per unit area,  $q$ , (in  $\text{g ha}^{-1} \text{y}^{-1}$ ), the soil cadmium surface density,  $C$ , is in the steady state given by

$$C'_S{}^* = qG'_S$$

But from equation (5)

$$C_P^* = P_{SP}C_S^*$$

and

$$C'_S{}^* = C_S^* d\sigma$$

Therefore

$$P''_{SP} = C_P^*/C'_S{}^* = P_{SP}C'_S{}^*/(d\sigma)/C'_S{}^* = P_{SP}/d\sigma \quad (6)$$

and

$$sP''_{OP} = C_P^*/q = \frac{\beta S\alpha\sigma(\sigma + \theta\alpha)d}{d\sigma(\sigma + \theta\alpha)\alpha(ST + L)}$$

or

$$sP''_{OP} = C_P^*/q = \frac{\beta S}{(ST + L)} \quad (7)$$

Thus, in the steady-state, the concentration in plants and the application rate are related by an expression which is independent of  $\alpha$ .

The fraction of a given input to soil or the fractional steady-state output flux from crops is simply given by the partitive coefficient,  $W_{SC}$ , where

$$W_{SC} = ST/(ST + L) \quad (8)$$

The effects of recirculation

When a fraction,  $\gamma$ , is recirculated from plants to soil particulates in organic residues and plant wastes, the transpiration rate is effectively reduced to  $T(1 - \gamma)$ . The expressions of particular interest then become:

$$G'_S = \frac{(\sigma + \theta\alpha)d}{\alpha(ST(1 - \gamma) + L)}$$

$$P'_{SP} = \frac{\beta S \alpha \sigma}{(\sigma + \theta\alpha)} \quad (\text{i.e. unchanged})$$

$$sP''_{OP} = \frac{\beta S}{(ST(1 - \gamma) + L)}$$

In this case, the partitive coefficient,  $W_{SC}$ , is given by the expression

$$W_{SC} = \frac{ST(1 - \gamma)}{ST(1 - \gamma) + L}$$

so that its value is smaller when there is recirculation than when there is no recirculation.

The superposition principle and multiple sources of contamination

If the plant concentration at the start of a forecasting period is  $C_P^\circ$ , corresponding to a concentration  $C_S^\circ$  in soils, and the future rate of application of a chemical contaminant is  $q$ , and is constant and the only source of contamination, the concentration in plants at time,  $t$ , later is

$$C_P(t) = (q sP''_{OP} - C_P^\circ)(1 - \exp(-t/G_S)) + C_P^\circ$$

$$\text{i.e. } C_P(t) = C_P^* - (C_P^* - C_P^\circ) \exp(-t/G_S)$$

or

$$C_P(t) = C_P^* (1 - \exp - t/G_S) + C_P^0 \exp - t/G_S$$

This last expression can be viewed as the sum of two contributions. The first is the concentration in plants, following the action of the application rate,  $q$ , over a time period,  $t$ . The second is the decay of the initial concentration,  $C_P^0$ , corresponding to the gradual loss of the initial cadmium input to soils.

#### Rates of change of concentrations in soil and plants

The rate of change of the concentration in soils, expressed in terms of the initial soil concentration,  $C_S^0$ , can be readily derived as follows

$$C_S(t) = C_S^* (1 - \exp - t/G_S) + C_S^0 \exp - t/G_S$$

$$\frac{dC_S}{dt} = C_S^* (1/G_S) \exp - t/G_S - C_S^0 (1/G_S) \exp - t/G_S$$

$$(1/C_S^0) \frac{dC_S}{dt} = (1/G_S) \exp - t/G_S (C_S^*/C_S^0 - 1)$$

At  $t = 0$

$$(1/C_S^0) \frac{dC_S}{dt} (0) = (1/G_S) (C_S^*/C_S^0 - 1)$$

But

$$C_S^* = q_m G_S \quad \text{so that} \quad (1/C_S^0) \frac{dC_S}{dt} (0) = (q_m/C_S^0 - 1/G_S)$$

The rate of change of plant concentrations is given by the expression

$$(1/C_P^0) \frac{dC_P}{dt} (0) = (1/G_S) (C_P^*/C_P^0 - 1)$$

### Natural source contributions

If the condition of soils and plants has been influenced by a natural source component,  $q_n$ , it can reasonably be assumed that the corresponding concentrations in soil and plants have already reached equilibrium. These contributions can be represented by  $C_S^n$  and  $C_P^n$ . They are assumed to persist in the future.

The general formulae for future soil and plant concentrations then become

$$C_S(t) = C_S^* (1 - \exp - t/G_S) + (C_S^o - C_S^n) \exp - t/G_S + C_S^n$$

$$C_P(t) = C_P^* (1 - \exp - t/G_S) + (C_P^o - C_P^n) \exp - t/G_S + C_P^n$$

where  $C_S^*$  and  $C_P^*$  are the steady-state concentrations in soils and plants corresponding to sources,  $q$ , which do not include the natural source  $q_n$ . These formulae imply that in general

$$C_S^o \geq C_S^n \quad \text{and} \quad C_P^o \geq C_P^n$$

so that present concentrations reflect some contamination additional to that caused by natural sources acting in isolation.

### Interpretation of transfer coefficients

The mean residence time for cadmium in soil defines the relationship between the steady-state burden in soils and the rate of application of cadmium to such soils. For example, if

$$q = 2 \text{ g ha}^{-1} \text{ y}^{-1} \quad \text{and} \quad G'_S = 100 \text{ y}$$

$$\text{then } C'_S = 200 \text{ g ha}^{-1}$$

The transfer coefficient  $P''_{SP}$  defines a relationship between the steady-state burden in soil per unit surface area and the steady-state concentration in plants. For example, if

$$C_S = 200 \text{ g ha}^{-1} \quad \text{and} \quad P''_{SP} = 0.9 \text{ } \mu\text{g g}^{-1} \text{ per kg ha}^{-1}$$

$$\text{then } C_P^* = 0.18 \text{ } \mu\text{g g}^{-1}$$

The transfer coefficient  $sP''_{OP}$  defines a relationship between the steady-state concentration in plants and the rate of application of cadmium to soil. For example, if

$$q = 2 \text{ g ha}^{-1} \text{ y}^{-1} \quad \text{and} \quad sP''_{OP} = 8 \text{ } \mu\text{g g}^{-1} \text{ per kg ha}^{-1} \text{ y}^{-1}$$

$$\text{then } C_P^* = 0.016 \text{ } \mu\text{g g}^{-1}$$

In practice, the mean residence time for soils,  $G'_S$ , can also be used to forecast the gradual accumulation of cadmium in soils under a constant rate of application. The transient concentration in soil,  $C_S(t)$ , will be given by the expression

$$C_S(t) = C_S^* (1 - \exp(-t/G'_S)) = G'_S q (1 - \exp(-t/G'_S))$$

Concentrations in plants will follow those in soils so that the time series for plants can be obtained from  $C_S(t)$  by multiplying by the appropriate transfer coefficient

$$C_P(t) = C_P^* (1 - \exp(-t/G'_S)) = q \cdot sP''_{OP} (1 - \exp(-t/G'_S))$$

The transfer coefficient,  $sP''_{OP}$ , also defines a relationship between a finite input to soil,  $Q$ , and the consequential integrated concentration or exposure commitment,  $E_P$ , to plants. For example,

$$Q = 5 \text{ kg ha}^{-1}$$

$$sP_{OP} = 50 \text{ } \mu\text{g g}^{-1} \text{ per kg ha}^{-1} \text{ y}^{-1}$$

$$= 50 \text{ } \mu\text{g y g}^{-1} \text{ per kg ha}^{-1}$$

so that

$$E_P = 250 \text{ } \mu\text{g y g}^{-1}$$

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