

# OECD PROCEEDINGS

## *Sources of Cadmium in the Environment*

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ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

## **Cadmium in Europe: Sources, Environmental Levels, Exposure to Humans**

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### **1. Introduction and overview**

This paper is prepared by Malcolm Hutton of Environmental Resources Management (ERM). It provides an overview of cadmium in Europe for the following topics:

- Emissions to atmosphere
- Inputs to landfill
- Inputs to surface waters
- Atmospheric concentrations
- Deposition from the atmosphere
- The application of phosphate and other fertilisers to soil
- Concentrations in soil
- Concentrations in rivers - water and sediments
- Concentrations in food
- Daily intake from food and other sources
- Tolerable daily intake

In addition, an annex [**not reproduced in this OECD publication**] provides certain data on emission of cadmium from the following sectors:

- non-ferrous metal production
- iron and steel production
- fossil fuel combustion

The paper's objective is to provide a broad range of information which can be used to place the issue of cadmium in Europe into perspective.

## 2. Emissions to the atmosphere

Hutton (1982) has assessed the sources of, human exposure to, and environmental impact of cadmium in the European Community. At that time the member states were: France, West Germany (FRG), Italy, the Netherlands, Belgium, the UK, Luxembourg and Ireland.

He made estimates of the atmospheric emissions from the major sources of cadmium for the year 1979. For most of the sources, he gives the emissions for each individual state as well as the total (all states) emissions. A summary of his results is presented in Annex B (Table B1),<sup>1</sup> and the national totals from these results are given in Table 1.

This study appears to be one of only two emissions inventories for member states carried out by a single author, or group of authors; the second one is the ESQUAD project - mentioned later. Several inventories for individual states have been reported, but comparisons between them must be viewed with caution, since the authors do not always use the same source categories or methods of estimation, and the years for which the estimates are made are usually different.

Ros and Slooff (1988) have summarised some of the estimates of atmospheric emissions of cadmium reported in the literature from the late 1970s onwards, and have also estimated the emissions in the Netherlands for the year 1985. These estimates are contained in Tables 2.15 and 2.16 in their report; Table 2.15 and that part of Table 2.16 dealing with atmospheric emissions are given in Annex B of this report (Tables A2 and A3).

If the national totals from Ros and Slooff's Table 2.15 are combined with the total for the Netherlands implied in their Table 2.16, then the summary of their figures given in Table 1 below results.

Environmental Resources Management have also made a summary of various estimates of cadmium emissions (ERL, 1990) and their values are listed in Table 1

Finally, the ESQUAD project has compiled and published an emissions inventory for cadmium and other pollutants (ESQUAD, 1994). Their values are also given in Table 1.

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<sup>1</sup> The Annex is not reproduced in this OECD publication.

**Table 1**

**Annual atmospheric emissions of cadmium from various sources, in various years, made by various authors, for seven EU states**

| Author               | Year    | Total cadmium emissions (t y <sup>-1</sup> ) |      |      |      |     |      |       |
|----------------------|---------|--|------|------|------|-----|------|-------|
|                      |         | NL   | UK   | FRG  | Belg | Dk  | Fr   | Italy |
| Hutton, 1982*        | 1979/80 | 3.6  | 15.8 | 29.2 | 2.5  | 4.3 | 19.6 | 14.8  |
| Ros and Slooff, 1988 |         | 4.1  | 14   | 33   | 22   | 5   | 29   | 21    |
| ERL, 1990            |         | 6.5  | 26.4 | 44.9 | 9.3  | 2.9 | 29.1 | 17.9  |
| ESQUAD, 1994         | 1990    | 5  | 52   | 74** | 18   | 2   | 46   | 49    |

\* Only the five major sources are included, see Table A1. \*\* The FRG and DDR combined

There are both agreements and disagreements within these four sets of estimates, but the overall agreement is poor. The different years for which the estimates were made presumably accounts for some, but not all of the discrepancies. The different methods used by the various authors concerned are probably a more important factor.

The difficulties in using the estimates made by different authors to compare the emissions from different countries are exemplified by a comparison of four sets of estimates of cadmium emissions for the UK. There are substantial discrepancies between the estimates, largely as a result of the different emission factors used. A summary of the estimates is given in Annex B, Table A4.

It might be argued that a comparison of national emissions of the kind suggested by Table 1 is too simple in that it takes no account of the size of the countries concerned. The areas of the seven member states being considered are given in Table 2.

**Table 2**

**Total areas of seven EU states (OECD, 1993)**

| Total area, 1000 km <sup>2</sup> |     |     |      |    |     |       |
|----------------------------------|-----|-----|------|----|-----|-------|
| NL                               | UK  | FRG | Belg | Dk | Fr  | Italy |
| 37                               | 245 | 249 | 31   | 43 | 552 | 301   |

If the cadmium emissions given, for example, by Ros and Slooff as in Table 1, are divided by the respective areas of the countries concerned, then the emissions per unit area of each country are obtained. These are given in Table 3.

**Table 3**

**Annual atmospheric emissions of cadmium per unit area of the country,  
as derived from Ros and Slooff's emission data and Table 2**

| NL   | UK   | FRG  | Belg | Dk   | Fr   | Italy |
|------|------|------|------|------|------|-------|
| 0.11 | 0.06 | 0.13 | 0.71 | 0.12 | 0.05 | 0.07  |

### 3. Inputs to landfill

The cadmium input to land is one of the more difficult parameters to quantify in this assessment.

Some of the individual land compartments - e.g. landfill and agricultural land - are apparently easy to define, but there are some problems. Should storage of jarosite on the factory premises, or the use of phosphorous slag for road foundations, be classed under the general heading of "landfill"? How important is flytipping? As regards agricultural land, is total deposition or deposition per unit area the more important parameter?

There are sometimes differences in the source categories used by different authors which make it difficult or impossible to compare their inventories.

Finally, for any one country, the sum of all inputs to all land compartments is difficult to estimate because of the problem of double counting.

In order to minimize these problems, but still allow some comparisons between countries, just one compartment - landfill - is considered in this section; it will be taken to include the storage of industrial waste on factory premises but not flytipping. Estimates of the inputs to another important compartment - agricultural land - are given in a subsequent section (the application of phosphate and other fertilizers to soil).

Hutton (1982) has made estimates of the cadmium inputs to land, for various source categories. The estimates were for the European Community as a whole, but for some sources the figures for individual states were given. His estimates for those sources not involving the problem of double counting, and which seem to apply to landfill, are given in Annex B (Table A5).

Ros and Slooff (1988) give data for the Netherlands. Their estimates for industrial waste are given in their Table 2.13. Their estimate for household refuse is about 33 tonnes per year (their Figure 2.2). A summary of their estimates is given in Annex B (Table A6).

There are clearly some discrepancies between the estimates given by the three sets of authors. However, the figures suggest that inputs of cadmium to landfill are higher for the UK and FRG than most other member states.

The most complete inventory of cadmium inputs to land in the EU member states appears to be that carried out by Environmental Resources Management (ERL, 1990). The estimates of cadmium inputs to landfill inferred from the ERL report - by country and activity - are summarised in Annex B (Table B7).

The estimates of cadmium inputs (all-source totals) to landfill from each of these three inventories are given in Table 4.

**Table 4**

**Estimates of cadmium inputs to landfill; various authors**

| Author                    | Cadmium input (t y <sup>-1</sup> ) |     |     |      |    |     |      |
|---------------------------|------------------------------------|-----|-----|------|----|-----|------|
|                           | NL                                 | UK  | FRG | Belg | Dk | Fr  | Ital |
| Hutton, 1982 <sup>a</sup> | 72                                 | 566 | 474 | 99   | 7  | 279 | 287  |
| ERL, 1990 <sup>b</sup>    | 94                                 | 274 | 336 | 87   | 32 | 230 | 201  |
| ERL, 1990 <sup>c</sup>    | 148                                | 336 | 473 | 169  | 35 | 343 | 311  |
| Ros and Slooff, 1988      | 78                                 | -   | -   | -    | -  | -   | -    |

*a* For only 4 source categories; see Annex B, Table B5.

*b* For the same 4 sources as given for Hutton.

*c* For all sources - as listed in Annex B, Table B7.

#### 4. Inputs to surface waters

The only country-by-country inventory of inputs of cadmium to water for the EU appears to be that carried out by Environmental Resources Management (ERL, 1990). They considered that only four sources made significant inputs to water. One of these, phosphate fertilizer manufacture, was important for only two countries: France with 10 tonnes of cadmium per year to the Seine estuary, and the Netherlands with 20 tonnes (cadmium per year), stated to be disposed of by dumping at sea. For both, landfill alternatives were being sought, and neither is included in Table 5 below.

The values in Table 5 indicate that inputs of cadmium to surface waters from industrial sources excluding phosphate fertilizer manufacture are lower for Denmark and the Netherlands than most other member states.

Table 5

**Inputs of cadmium (tonnes per year) to surface waters from industrial sources excluding phosphate fertilizer manufacture; adapted from ERL, 1990**

| Source                   | Cadmium input (t y <sup>-1</sup> ) |      |      |      |     |      |      |
|--------------------------|------------------------------------|------|------|------|-----|------|------|
|                          | NL                                 | UK   | FRG  | Belg | Dk  | Fr   | Ital |
| Iron and steel           | 0.9                                | 3.0  | 6.3  | 2.4  | 0.1 | 3.0  | 3.6  |
| Refining, non-ferrous    | 1.4                                | 4.5  | 7.6  | 2.5  | 0.0 | 5.2  | 3.4  |
| Cd processing industries | 0.1                                | 18.9 | 12.3 | 0.1  | 0.1 | 12.4 | 0.9  |
| Total                    | 2.4                                | 25.4 | 26.2 | 5.0  | 0.2 | 20.6 | 7.9  |

The inputs of cadmium to surface waters in the Netherlands are also considered in a recent Dutch document (SPEED, 1993). This report implies that gypsum waste from the phosphate fertilizer industry (estimated to contain 14.5 tonnes of cadmium in 1985) is discharged to surface waters rather than to the sea. However, it is stated that the cadmium input from this source is expected to decline to 1.2 tonnes in 1994, and it is planned to reduce it to zero by the year 2000. The SPEED report estimates the input from the other industrial sources in 1985 to be 1.1 tonnes per year - significantly less than the ERL figure. Estimates are also given in the SPEED report of the inputs from *inter alia* domestic waste water (0.7 t y<sup>-1</sup>), aerial deposition (about 1 t y<sup>-1</sup>), agriculture and traffic (each about 0.1 t y<sup>-1</sup>). Their estimate of the total for 1985 is about 18.5 t<sup>-1</sup>, of which 14.5 t y<sup>-1</sup> is from the fertilizer industry. The ranking of the Netherlands as regards cadmium inputs to surface waters is clearly very dependent on whether or not waste gypsum is included.

In any assessment of inputs of cadmium to surface waters in the Netherlands, the contributions from abroad must be considered. In the SPEED report, it is estimated that in 1985 about 11 tonnes were brought in by the Rhine and the Meuse, and that even smaller rivers such as the Dommel and the Roer brought in about 2 tonnes. A fuller presentation of the transboundary flows of cadmium into the Netherlands is given in another Dutch report (XI/310/90-EN). Table 6 is taken from that report.

As is noted in the report, since the beginning of the 1980s there has been a fall in the amount of cadmium carried in each of the three rivers flowing across the border. The situation in the Meuse apparently started to deteriorate again in 1986, perhaps because a new metal-processing plant was established in Ehein (Belgium). However, as a result of subsequent Dutch/Belgian discussions, measures have been taken which seem to have had some effect, to judge by the lower levels in 1989.

Table 6

**Cadmium entering the Netherlands via the Rhine, the Meuse and the Schelde  
in tonnes per year (Report XI/310/90-EN)**

| River            | Cadmium entering the Netherlands, t <sup>-1</sup> |      |      |      |      |      |      |
|------------------|---|------|------|------|------|------|------|
|                  | 1975  | 1980 | 1985 | 1986 | 1987 | 1988 | 1989 |
| Rhine (Lobith)   | 132   | 117  | 9    | 12   | 10   | 10   | 6    |
| Maas (Eijsden)   | 19  | 37   | 3    | 3    | 14   | 20   | 3    |
| Scheldt (Schaar) | 16  | 11   | 8    | 3    | 5    | 5    | ns   |
| Total            | 167   | 165  | 20   | 18   | 29   | 35   | ns   |

*ns = not stated*

## 5. Atmospheric concentrations

The WHO, in a review of airborne cadmium levels in Europe (WHO, 1987), found that annual means in rural areas ranged from <1 to 5 ng m<sup>-3</sup>, compared with 5-15 ng m<sup>-3</sup> in urban areas, and 15-50 ng m<sup>-3</sup> in industrialised areas. Most of the references used in the review date from the early 1970s, and there is some evidence that levels have declined during the past two decades, so it is likely that current levels are somewhat lower than the values given in the review. However, there are more recent data.

Ros and Slooff (1988) give a considerable body of information on airborne cadmium levels in the Netherlands. In introducing these data they remark In comparison to data from abroad the values reported in the Netherlands are not high. The large-scale annual average cadmium concentrations over the Netherlands (for the period 1982/83) have been derived from measured values combined with a distribution model. The concentrations ranged from about 1.8 ng m<sup>-3</sup> in the south to about 0.5 ng m<sup>-3</sup> in the north. The authors also report the measured, long-term average concentrations at various specific locations (their Tables 4.8, 4.9 and 4.10), found during the period 1975 to 1983; most of the measurements were made during 1980/83. The highest single value was 11 ng m<sup>-3</sup>, in an industrial area - Nijmegen, Waalhaven. None of the others was greater than 6 ng m<sup>-3</sup>, with most values in the range 0.7 to 3 ng m<sup>-3</sup>. Ros and Slooff comment that no trends with time could be seen in the period 1975/83.

Ros and Slooff cite a German study in which the annual averages (1981-1982) in four large cities and one rural site were reported to be in the range 1 to 3 ng m<sup>-3</sup>. In another German study cited by Lahmann et al (1986), the concentrations of cadmium and other metals were measured at 21 sites in Berlin during the year 1982/83; the concentrations of cadmium in trafficked and residential areas were much the same - 4.0 and 3.8 ng m<sup>-3</sup> respectively. In a third German study, cited by Jensen and Bro-Rasmussen

(1992), measurements made in Frankfurt during the period 1973 to 1985 showed a downward trend from about  $10 \text{ ng m}^{-3}$  in 1973/74 to  $2\text{-}3 \text{ ng m}^{-3}$  in 1985.

Lee et al (1993) have analysed and presented data from several long-term studies of trace element concentrations in the UK. Their results, some of which are presented in Table 7, show a rural/urban/industrial gradient. They also indicate a significant decrease of cadmium concentration with time at all three types of site.

**Table 7**

**Concentrations of cadmium in air at various sites in the UK (Lee et al, 1993)**

| Type and number of sites | Mean cadmium concentration, $\text{ng m}^{-3}$ |         |
|--------------------------|--|---------|
|                          | 1975-78  | 1986-89 |
| Rural (2)                | 1.92   | 0.76    |
| Urban (6)                | 7.8  | 2.0     |
| Industrial (1)           | 33   | 21      |

The authors do not give the values of the mean annual concentrations for each year in tabular form. However, it is evident from their graphical presentations that the concentrations in the period 1980-83 (the period in which most of the Dutch measurements reported by Ros and Slooff were made) were closer to the 1975-78 values in Table 4 than to the 1986-89 ones.

In another set of measurements made at a rural location in the UK, Yaaqub et al reported an annual average concentration of  $1.1 \text{ ng m}^{-3}$  for the year 1987/88. In a survey of heavy metal concentrations around a smelter in Avonmouth, UK, Davis and Clayton (1985) found 3-monthly average cadmium concentrations down wind of the works ranging from  $80 \text{ ng m}^{-3}$  (at 500 m) to  $10 \text{ ng m}^{-3}$  (at 2 km).

Some of the cadmium-in-air concentrations described above are summarised in Table 8.

There are, then, useful data on the levels of airborne cadmium in the Netherlands, Germany and the UK. They suggest that - during the early 1980s - the levels in the Netherlands and Germany were about the same, and that those in the UK were rather higher. What other data there are for EU states (e.g., Jansen and Bro-Rasmussen, 1992) do not provide any evidence that cadmium in air concentrations is significantly higher in particular states.

Table 8

Concentrations of airborne cadmium ( $\text{ng m}^{-3}$ ) in three member states

| Country                       | Year    | Airborne cadmium<br>$\text{ng m}^{-3}$ | Reference                      |
|-------------------------------|---------|--|--------------------------------|
| <b>The Netherlands</b>        |         |  |                                |
| South (overall mean)          | 1982/83 | 1.8                                    | Ros and Slooff, 1988           |
| North (overall mean)          | 1982/83 | 0.5                                    | Ros and Slooff, 1988           |
| Maastricht (urban/industrial) | 1975/81 | 5 (median)                             | Ros and Slooff, 1988           |
| Gelderland (urban/industrial) | 1980/83 | 3-11                                   | Ros and Slooff, 1988           |
| Southern areas (urban/rural)  | 1980/83 | 1-3                                    | Ros and Slooff, 1988           |
| <b>FRG</b>                    |         |  |                                |
| Four cities, one rural site   | 1981/82 | 1-3                                    | Ros and Slooff, 1988           |
| Berlin                        | 1982/83 | 4                                      | Lahmann et al. 1986            |
| Frankfurt                     | 1973/74 | 10                                     | Jansen and Bro-Rasmussen, 1992 |
| Frankfurt                     | 1985    | 2-3                                    | Jansen and Bro-Rasmussen, 1992 |
| <b>UK</b>                     |         |  |                                |
| Two rural sites               | 1975/78 | 1.9                                    | Lee et al, 1993                |
| Two rural sites               | 1986/89 | 0.8                                    | Lee et al, 1993                |
| Six urban sites               | 1975/76 | 8                                      | Lee et al, 1993                |
| Six urban sites               | 1986/89 | 2                                      | Lee et al. 1993                |
| One industrial site           | 1975/78 | 33                                     | Lee et al. 1993                |
| One industrial site           | 1986/89 | 21                                     | Lee et al. 1993                |

## 6. Deposition from the atmosphere

Various processes contribute to the deposition of suspended material from the atmosphere.

Dry deposition may occur in three ways: diffusion, gravitational settling, and downward turbulent transport followed by retention at the ground as a result of processes such as impaction and absorption. The controlling parameter is the aerodynamic diameter of the particles, and the last of these three ways is usually the major one for atmospheric cadmium.

Wet deposition or washout is clearly a function of the rainfall in the area concerned. For an annual rainfall in the range 500 to 1000 mm, the wet deposition is one to two times the dry deposition.

Total deposition is the quantity of prime interest in this report in that it is the measure of total aerial deposition to soil. Unless otherwise stated, the term deposition will be used with the meaning of total deposition.

In summary then, the rate of deposition of suspended material from the atmosphere depends upon: the concentration of the material, its size distribution and the amount of rainfall.

It is possible to make an estimate of the rate of deposition of a suspended particulate material (of known concentration and size distribution) if the annual rainfall is known. For metal particles formed by combustion processes, and an annual rainfall of about 600 mm, an airborne concentration of  $1 \text{ ng m}^{-3}$  would give rise to a total deposition rate of about  $1.5 \text{ g ha}^{-1} \text{ y}^{-1}$  (WHO, 1987). If the average rural cadmium concentration in the EU is taken to be  $1.5 \text{ ng m}^{-3}$ , it follows that the corresponding deposition rate would be about  $2 \text{ g ha}^{-1} \text{ y}^{-1}$ .

Some of the reported measurements of deposited airborne cadmium found in the literature are summarised in Table 9.

In the Netherlands, Ros and Slooff (1988) report national averages (presumably rural and urban combined) of  $2.6$  and  $1.4 \text{ g ha}^{-1} \text{ y}^{-1}$  for the years 1983 and 1985 respectively. The mean of these,  $2 \text{ g ha}^{-1} \text{ y}^{-1}$ , is about the same as the value estimated assuming a national mean air concentration of  $1.5 \text{ ng m}^{-3}$ . More recent data (CCR, 1991) show a decline in the national average deposition rate during the period 1983-1990, with a value of about  $1 \text{ g ha}^{-1} \text{ y}^{-1}$  in 1990.

Ros and Slooff (1988) also report annual concentrations of cadmium in rainfall. They give the annual time-weighted average concentrations at 31 locations over the Netherlands for three different years. These concentrations are a measure of total cadmium deposition, and with the assumption of a value for the average annual rainfall of about 600 mm, they can be translated into  $\text{g ha}^{-1} \text{ y}^{-1}$ . The results are presented in Table 10.

Ros and Slooff comment that the highest values were observed near Eindhoven - presumably because of the proximity of the Budelco plant, and Belgian metal works.

It may be concluded from Table 9 that the deposition rates of cadmium in rural locations are similar in different EU member states.

**Table 9**  
**Measured total deposition rates**

| Location           | Deposition<br>$\text{g ha}^{-1} \text{y}^{-1}$ | Comments                               | Reference                      |
|--------------------|--|--|--------------------------------|
| <b>Rural</b>       |  |  |                                |
| EC states          | 3  | Representative value for rural soils   | Hutton, 1982                   |
| EC states          | 0.05-3   | Remote areas                           | Lahmann et al, 1986            |
| EC states          | 0.5-25   | Other rural areas                      | Lahmann et al, 1986            |
| Denmark            | 2  | Mean national value 1975-78            | Jensen and Bro-Rasmussen, 1992 |
| Denmark            | 0.9  | Mean national value 1988               | Jensen and Bro-Rasmussen, 1992 |
| FRG                | 1-4  | Mean values, 1980-84                   | Jensen and Bro-Rasmussen, 1992 |
| Lancashire, UK     | 1.7  | A single site, 1978/79                 | Harrison and Williams, 1982    |
| <b>Urban</b>       |  |  |                                |
| EC states          | 1.5-100  |  | Lahmann et al, 1986            |
| Essen and Dortmund | 8  | 1980/81                                | Jensen and Bro-Rasmussen, 1992 |
| Essen and Dortmund | 3-5  | 1983/84                                | Jensen and Bro-Rasmussen, 1992 |
| Walsall, UK        | 120  | Industrial town, 1980                  | Cawse, 1982                    |
| Inner London       | 25   | Mean of two sites                      | Harrison et al, 1975           |
| Inner London       | 7  | Mean of three sites                    | Duggan and Burton, 1983        |
| North London       | 18   | Downwind of a major refuse incinerator | Hutton and Wadge, 1988         |

\*Dry deposition only

**Table 10**

**Cadmium in rainfall in Holland, and the corresponding estimated cadmium deposition rate per unit area (derived from Ros and Slooff, 1988)**

| Year | Cd in rainfall mg L <sup>-1</sup> | Cd total deposition rate g ha <sup>-1</sup> y <sup>-1</sup> |
|------|-----------------------------------|---|
| 1981 | 0.10-0.72                         | 0.6-4.3   |
| 1982 | 0.17-0.75                         | 1.0-4.5   |
| 1985 | 0.10-0.4                          | 0.6-2.4   |

In the Dutch Policy on Cadmium document (Alders, 1991), it is stated that 80% of the cadmium deposition in the Netherlands comes from abroad. The justification for this statement is not clear. It is true that the deposition rates implied by Table 3 (the figures in the table should be divided by 10 to convert them into units of g ha<sup>-1</sup> y<sup>-1</sup>) are much lower than the measured values given in Table 8. There is no obvious explanation for this discrepancy. However, it appears to exist for all countries - not just Holland - and does not provide a basis for estimating transboundary cadmium deposition.

## 7. The application of phosphate and other fertilizers to soil

### 7.1 Countrywide deposition

Whereas aeriaily deposited cadmium falls on all outdoor surfaces, cadmium in fertilizers is deposited only on agricultural land. It is not always easy to determine the area of agricultural land, in any one state, which is treated with a particular type of fertilizer. Consequently, the figure for the total (national) annual deposition of cadmium in fertilizers to soil is not always readily translatable into a deposition rate per hectare.

Hutton (1982) has made estimates of the annual deposition rates of cadmium to agricultural land from the application of phosphate fertilizers in the EC for the year 1979. More recent estimates have been reported for the Netherlands (Ros and Slooff, 1988), for all the member states (ERL, 1990), and for some of the states (OECD, 1994). These data are summarised in Table 11

**Table 11**

**Annual deposition of cadmium to agricultural land from the use of phosphate fertilizers. The estimates of various authors**

| Reference            | Year  | Cadmium deposition (t y <sup>-1</sup> ) |    |     |      |    |     |       |
|----------------------|-------|---|----|-----|------|----|-----|-------|
|                      |       | NL                                      | UK | FRG | Belg | Dk | Fr  | Italy |
| Hutton, 1982         | 1979  | 7                                       | 45 | 44  | 8    | 7  | 144 | 49    |
| Ros and Slooff, 1988 | 1985  | 7                                       | -  | -   | -    | -  | -   | -     |
| ERL, 1990            |       | 5                                       | 25 | 40  | 7    | 6  | 82  | 44    |
| OECD, 1994           | 1990* | 3                                       | -  | 25  | 6    | -  | -   | -     |

\*1990 for the Netherlands, 1991 for Germany, not stated for Belgium.

There are other fertilizer materials which contain significant concentrations of cadmium, but data as to their cadmium content and the extent of their use are usually less complete than for phosphate fertilizers.

The materials of concern are:

- Fertilizers without phosphates (calcium, nitrogen and potassium fertilizers).
- Animal manure (from animals fed on fodder phosphate).
- Sewage sludge.

Fertilizers without phosphates are a relatively minor contributor to soil cadmium. Ros and Slooff (1988) estimate that they deposited only about 0.4 tonnes of cadmium to Dutch soil in 1985 - compared with 7 tonnes from phosphate fertilizers. For Belgium, the OECD (1994) give corresponding figures of 1 and 6 tonnes, respectively.

Animal manure is likely to be an important source. For the year 1990, the OECD (1994) give estimates of 4.5 and 2 tonnes of cadmium deposited to soil for Holland and Belgium respectively. No estimates for any other states have been found.

Estimates of the quantity of cadmium disposed of by the use of sewage sludge on agricultural land for the EU have been published by ERL (1990). Jensen and Bro-Rasmussen (1992) give some figures for Denmark and the Netherlands, and Ros and Slooff (1988) also give an estimate - much lower - for the Netherlands. Additionally, there are some fairly recent data for the UK (DoE, 1993) and Germany (OECD, 1994). Table 12 gives a summary of these estimates.

Table 12

**Annual deposition of cadmium to agricultural land from the use of sludge.  
The estimates of various authors**

| Reference                         | Year | Cadmium deposited $t\ y^{-1}$ |      |      |     |     |       |
|-----------------------------------|------|-------------------------------|------|------|-----|-----|-------|
|                                   |      | NL                            | UK   | FRG  | Dk  | Fr  | Italy |
| Ros and Slooff, 1988              | 1985 | 0.8                           | -    | -    | -   | -   | -     |
| ERL, 1990                         |      | 3.2                           | 16.6 | 19.7 | 0.4 | 5.8 | 5.5   |
| Jensen and Bro-Rasmussen,<br>1992 |      | 3.2                           | -    | -    | 0.4 | -   | -     |
| OECD, 1994                        | 1991 | -                             | -    | 2.4  | -   | -   | -     |
| DoE, 1993                         | 1991 | -                             | 2.3  | -    | -   | -   | -     |

Because of the wide differences in the values given in Table 12, it is difficult to make reliable estimates of total, national deposition rates of cadmium in sewage sludge.

### 7.2 Deposition per unit area of soil

Hutton (1982) has derived figures for the annual input of cadmium (from phosphate fertilizer) per hectare of arable land, from estimates of the rate of fertilizer input and of the cadmium content of the fertilizer. His results are presented in Table 13. It should be noted that the data for annual fertilizer input, on which these estimates are based, are for the year 1967/68.

Table 13

**Estimates of the rates of cadmium input, per unit area, to arable land  
arising from the use of phosphate fertilizer (Hutton, 1982)**

| Cadmium input to soil ( $g\ ha^{-1}\ y^{-1}$ ) |     |     |      |     |     |       |
|--|-----|-----|------|-----|-----|-------|
| NL   | UK  | FRG | Belg | Dk  | Fr  | Italy |
| 4.6  | 6.5 | 4.6 | 9.4  | 2.4 | 5.4 | 1.6*  |

\* Hutton comments that the figure for Italy may be unrealistic as fertilizer use doubled in the period 1967/68 to 1979/80.

There appear to be few other published estimates of the cadmium deposition rates per unit area resulting from the use of fertilizers. Jensen and Bro-Rasmussen (1992) p149 cite some work in which a figure for Denmark of  $2\ g\ ha^{-1}\ y^{-1}$  (arising from the use of phosphatic fertilizers) is given; this is close to the estimate in Table 13. The OECD (1994) pp123-125 quote cadmium deposition rates from "artificial fertilizers" for the Netherlands,

Germany and Denmark of 3.8, 3 and 3.2 g ha<sup>-1</sup> y<sup>-1</sup> respectively. These are all lower than the values given in Table 13.

If it is assumed that all the arable land in each state is treated with phosphate fertilizer, then an estimate of the deposition of cadmium per unit area from this source can be made from the national deposition data given in Table 11, and a knowledge of the area of arable and permanent crop land in each state. The latter data as given by the OECD (1993) for 1990 are shown in Table 14, and the resulting estimates of cadmium deposition per unit area in Table 15.

**Table 14**

**Areas of arable and permanent crop land in member states (OECD, 1993)**

| Area, 1000 km <sup>2</sup> |      |      |      |      |      |     |       |
|----------------------------|------|------|------|------|------|-----|-------|
| NL                         | UK   | W.Gy | E.Gy | Belg | Dk   | Fr  | Italy |
| 9.4                        | 66.7 | 74.9 | 49.2 | 7.6  | 25.7 | 192 | 120   |

The areas for both East and West Germany are given for completeness. The area for West Germany is used in the estimates given in Table 15.

**Table 15**

**Annual deposition of cadmium to arable land from the use of phosphate fertilizers. Estimated from the national input data in Table 10, and the areas given in Table 14**

| Reference    | Year | Cadmium deposition rate (g ha <sup>-1</sup> y <sup>-1</sup> ) |     |     |      |     |     |       |
|--------------|------|---|-----|-----|------|-----|-----|-------|
|              |      | NL  | UK  | FRG | Belg | Dk  | Fr  | Italy |
| Hutton, 1982 | 1979 | 7.4   | 6.7 | 5.9 | 10.5 | 2.7 | 7.5 | 4.1   |
| ERL, 1990    |      | 5.3   | 3.7 | 5.3 | 9.2  | 2.3 | 4.2 | 3.7   |

There is reasonable agreement between Table 15 and the earlier estimates of cadmium deposition per unit area from the use of phosphate fertilizer. This implies that the assumption that all arable land is treated with phosphate fertilizer is, for most states, not in serious error.

Detailed information about the use of sewage sludge on agricultural land is available for the UK (DoE, 1993). The cadmium content (about 5 g t<sup>-1</sup>) is roughly 10 times lower than that of phosphate fertilizer, but it is applied at a rate (about 9 t ha<sup>-1</sup>) roughly 100 times that for phosphate fertilizer. In 1991, only about 0.3 per cent of agricultural land in the UK was treated with sewage sludge, but the resulting input to this

land was a large one - about  $40 \text{ g ha}^{-1} \text{ y}^{-1}$ . No comparable data have been found for other states. Fraters and Beurden (1993) tabulate the cadmium load ( $\text{g ha}^{-1} \text{ y}^{-1}$ ) from the use of sewage sludge, for the EC member states (their Appendix II). However, the loadings they give are based upon the assumptions that the cadmium content of sewage sludge is uniform throughout the EU (at  $10 \text{ g Cd}$  per tonne of sludge), and that sewage sludge is applied uniformly to the total area of agricultural land in each member state. ERL also tabulate the cadmium load ( $\text{g ha}^{-1} \text{ y}^{-1}$ ) from the use of sewage sludge for each member state. However, like Fraters and Beurden, they assume a constant cadmium content of sewage sludge ( $23 \text{ g t}^{-1}$ ) for most of the member states; they also assume that, in each state, 1 per cent of agricultural land is treated with sewage sludge.

### 7.3 Conclusions

In the UK the deposition of cadmium, to arable soil, arising from the use of phosphate fertilizer is of greater significance at the national level than the agricultural application of animal manure or sewage sludge.

For other member states, it has not been possible to form reliable estimates of either the total or per unit area deposition rates of cadmium arising from the use of animal manure and sewage sludge.

## 8. Concentrations in soil

In a review of the natural occurrence of environmental cadmium, WHO (1992b) note that cadmium is widely distributed in the Earth's crust at an average concentration of about  $0.1 \text{ mg g}^{-1}$ , and that surface soil concentrations typically range from  $0.1$  to  $0.4 \text{ mg g}^{-1}$ .

Ros and Slooff (1988) give an account of the cadmium concentrations found in a survey of cultivated areas in the Netherlands. The samples were taken from the top 20 cm of soil, and grouped according to soil type. The summary given in Table 16 is from their report.

The authors make some comments as to the variation in concentration with geographical location. The concentrations in the northern provinces are somewhat lower ( $0.30 \text{ mg kg}^{-1}$ ) and those in the region of rivers somewhat higher ( $0.49 \text{ mg kg}^{-1}$ ) than concentrations in the rest of the country. For sandy soils, the values for the north-east of the country ( $0.31 \text{ mg kg}^{-1}$ ) and the polders of Lake IJssel ( $0.34 \text{ mg kg}^{-1}$ ) are about at an average level. Lower levels are found in the west ( $<0.2 \text{ mg kg}^{-1}$ ) and higher levels ( $0.41 \text{ mg kg}^{-1}$ ) in the south (Kempen).

**Table 16****Cadmium concentrations in cultivated soils in the Netherlands (Ros and Slooff, 1988)**

| Type of soil     | N*  | Cadmium in soil, mg kg <sup>-1</sup> | std devn |
|------------------|-----|--------------------------------------|----------|
| Clay soils       | 520 | 0.41                                 | 0.19     |
| Sandy soils      | 298 | 0.32                                 | 0.18     |
| Peat             | 40  | 0.87                                 | 0.32     |
| Cleared peatland | 43  | 0.30                                 | 0.19     |
| Loess            | 24  | 0.78                                 | 0.16     |
| TOTAL            | 925 | 0.40                                 | 0.19     |

\* the number of samples

These "higher levels in the south" are only marginally above the national average given in Table 16.

Jensen and Bro-Rasmussen have tabulated some values of cadmium concentrations found in normal agricultural soils in other member states, and these together with some additional values are given in Table 17.

It is evident from Table 17 that concentrations of cadmium in normal agricultural soils in EU Member states are relatively similar.

Cadmium concentrations in contaminated soils are, of course, likely to much higher than those in normal agricultural soils. For Dutch soils, Ros and Slooff (1988) quote levels of about 8 mg kg<sup>-1</sup> in samples from river forelands and polders raised with dredged material. Their report also gives in diagrammatic form the results of a survey made around the Budelco works, sampling the top 5 cm of soil. The cadmium concentrations along a NE line from the works at distances of 3, 15 and 20 km were about 8, 4 and 3 mg kg<sup>-1</sup> respectively.

Such concentrations in contaminated land are not high in comparison with reported values in other countries. The WHO (1992a) comment that soil cadmium concentrations in excess of 100 mg kg<sup>-1</sup> are commonly encountered close to long established smelters. In a survey of about 500 households in the UK village of Shipham (a former zinc mining area) the mean cadmium concentration in garden soils was 100 mg kg<sup>-1</sup> (Culbard et al, 1988).

## 9. Concentrations in rivers - water and sediments

### 9.1 Water

Cadmium in water exists as dissolved ion or complexes, or bound in colloids or particulate matter. The distinction between particulate bound and colloidal cadmium is arbitrarily set at a particle size of 0.4  $\mu\text{m}$  whereas the limit between dissolved and colloidal cadmium is around 0.01  $\mu\text{m}$ . In practice, however, distinction is only made between the fractions above and below 0.4  $\mu\text{m}$  ("dissolved" and "particulate" respectively) because of the technical difficulties associated with separation at 0.01  $\mu\text{m}$ . "Total" refers to dissolved plus particulate cadmium.

Ros and Slooff (1988) have given an account of the cadmium content of the major rivers in the Netherlands. Table 18a is adapted from their report.

Table 18a

Cadmium in water and in suspended matter, in three Dutch rivers  
(Ros and Slooff, 1988)

| River                           | Year | Cd concentration, $\text{mg L}^{-1}$ |       | Cd content of suspended matter, $\text{mg kg}^{-1}$ |
|---------------------------------|------|--------------------------------------|-------|---|
|                                 |      | dissolved                            | total |   |
| Rhine (Lobith)                  | 1976 | -                                    | 4.50* | -   |
|                                 | 1983 | 0.18                                 | 0.42  | 6.3   |
|                                 | 1984 | 0.059                                | 0.20  | 3.8   |
|                                 | 1985 | 0.034                                | 0.14  | 2.9   |
|                                 | 1986 | 0.026                                | 0.14  | 2.3   |
| Meuse (Eisjden)                 | 1976 | -                                    | 3.50* | -   |
|                                 | 1983 | 0.34                                 | 1.44  | 29  |
|                                 | 1984 | 0.072                                | 0.85  | 21  |
|                                 | 1985 | 0.045                                | 0.35  | 11  |
|                                 | 1986 | 0.059                                | 0.43  | 18  |
| Scheldt (Schaar van Ouden Doel) | 1976 | -                                    | 3.00* | -   |
|                                 | 1983 | 0.22                                 | 1.12  | 17  |
|                                 | 1984 | 0.18                                 | 1.29  | 16  |
|                                 | 1985 | 0.45                                 | 1.61  | 15  |
|                                 | 1986 | 0.31                                 | 0.96  | 9   |

\* Approximate value taken from their Figure 4.3

It can be seen that - with the possible exception of the Meuse - there is a pattern of decreasing values with time (for both total cadmium in water and cadmium in suspended matter).

More recent data are available from CCRX (1991), and these are given in Table 18b.

**Table 18b**

**Cadmium in suspended matter in three Dutch rivers (CCRX, 1991)**

| River                              | Year | Cd concentration mg kg <sup>-1</sup> |
|------------------------------------|------|--------------------------------------|
| Rhine (Lobith)                     | 1988 | 3.0                                  |
|                                    | 1989 | 3.5                                  |
|                                    | 1990 | 3.0                                  |
| Meuse (Eijsden)                    | 1988 | 133                                  |
|                                    | 1989 | 19                                   |
|                                    | 1990 | 32                                   |
| Scheldt (Westerschelde, Terneuzen) | 1988 | --                                   |
|                                    | 1989 | 2.1                                  |
|                                    | 1990 | 1.2                                  |

It is evident that cadmium concentrations in suspended matter in the Rhine have remained fairly constant since the mid-1980s, whereas those in the Meuse showed a sharp but temporary increase around 1988. This pattern corresponds with that of the inputs to the Meuse, already noted in Table 6.

The OECD (1993) have published total cadmium in water values for a number of European and other rivers. Table 19 is derived from their report.

Table 19 indicates a general decline in cadmium concentrations in most of the rivers since 1975.

Table 19

## Water quality of rivers, total cadmium (OECD, 1993)

| Location              | Cadmium concentration, mg L <sup>-1</sup> |       |      |                   |                                |
|-----------------------|---|-------|------|-------------------|--------------------------------|
|                       | 1975                                      | 1980  | 1985 | 1991 <sup>a</sup> | Avg, last 3 years <sup>b</sup> |
| <b>Netherlands</b>    |   |       |      |                   |                                |
| Meuse-Kaisersveer     | 0.90                                      | 1.50  | 0.21 | 0.18              | 0.38                           |
| Meuse-Eijsden         | 3.10                                      | 3.40  | 0.35 | 0.53              | 1.13                           |
| Nieuwe Waterweg(c)    | 1.00                                      | 0.90  | 0.26 | 0.16              | 0.18                           |
| Ijssel-Kampen         | 1.40                                      | 1.30  | 0.10 | 0.10              | -                              |
| Rhine-Lobith          | 2.30                                      | 1.60  | 0.14 | 0.11              | 0.11                           |
| <b>UK</b>             |   |       |      |                   |                                |
| Thames                | -   | 1.04  | 0.79 | 0.14              | 0.24                           |
| Severn                | 5.17                                      | 10.00 | 0.21 | 0.39              | 0.43                           |
| Clyde                 | 3.80                                      | 1.08  | 0.78 | 0.17              | 0.39                           |
| Mersey                | 20.00                                     | 0.79  | 0.19 | 0.30              | 0.26                           |
| <b>Germany</b>        |   |       |      |                   |                                |
| Rhine-Kleve-Bimmen    | -   | 1.40  | 0.30 | 0.10              | 0.17                           |
| Weser/Intschede       | -   | 0.50  | 0.50 | 0.50              | 0.50                           |
| Donau/Jochenstein (d) | -   | 0.20  | 0.10 | 0.30              | 0.30                           |
| <b>Belgium</b>        |   |       |      |                   |                                |
| Meuse-Heer/Agi(c)     | 0.80                                      | 0.30  | 0.22 | 0.21              | 0.34                           |
| Meuse-Lanaye(c)       | 2.60                                      | 1.20  | 0.39 | 1.02              | 2.00                           |
| Escaut-Doel(c)        | 1.50                                      | 5.80  | 1.22 | 0.82              | 0.92                           |
| <b>Luxembourg</b>     |   |       |      |                   |                                |
| Moselle               | -   | 4.10  | 2.00 | 0.10              | 0.50                           |
| Sûre(c)               | -   | 4.0   | 4.0  | 1.0               | -                              |
| <b>France</b>         |   |       |      |                   |                                |
| Rhin-Seltz(c)         | -   | 1.5   | 0.1  | 0.11              | -                              |
| <b>Italy</b>          |   |       |      |                   |                                |
| Po(e)                 | 0.16                                      | 0.05  | 0.12 | -                 | -                              |
| Adige(f,g)            | -   | 0.03  | 2.80 | 0.50              | -                              |
| Tevere                | -   | 0.09  | 0.02 | -                 | -                              |

## Notes

- (a) Data refer to 1990 or 1991.  
(b) The last three years after 1986 for which data available.  
(c) 1991 data refer to 1989.  
(d) 1991 data represent detection limits  
(e) 1975 data refer to 1977.  
(f) 1985 data refer to 1984.  
(g) 1991 data refer to 1988.

The increase in cadmium carried into Holland by the Meuse during 1987/88 and the subsequent decline (Table 6), and the corresponding rise and fall of the cadmium content of sediments (Table 18b), have already been noted. This pattern is entirely consistent with that of the cadmium concentrations in the Meuse shown in Table 19.

## 9.2 Sediments

Caution must be exercised in any comparison of the measured values (of cadmium in sediment) reported by different authors. Not all use the same extraction methods in the analytical procedures, and there is no uniformity as to the size fraction sampled. Some results refer to total sediments, others to the size fraction below 63 mm, or below 20 mm or below 16mm. Ros and Slooff (1988) give some of their results in terms of a "standard sludge" content, i.e. 50% of the calcium-free mineral particles below 16 mm.

These difficulties are usually absent in the comparison of results from the same author or group of authors, and such results are particularly valuable in the determination of trends with time.

Ros and Slooff (1988) tabulate cadmium concentrations found in dredged materials from various waters in and near Rotterdam. Table 20 is adapted from their report.

Table 20

### Cadmium in dredged sludge from various waters in and near Rotterdam (from Ros and Slooff, 1988)

| Harbour/canal       | Mean Cd content mg kg <sup>-1</sup> dw |         |
|---------------------|--|---------|
|                     | 1981                                   | 1985/86 |
| Waalhaven           | 19                                     | 10      |
| Eemhaven            | 12                                     | 6       |
| 1st Petroleum haven | 16                                     | 6       |
| Botlekhaven         | 15                                     | 7       |
| Exit Botlekhaven    | 9                                      | 3       |
| Caland Kanal        | 5                                      | ±1      |

The authors note that the data on harbour sludge (based simply on total dry weight) cannot be compared directly to the values for the canal, which are based on standard sludge; converted to standard sludge, the values for the harbour sludges will be considerably higher. However, it is clear that the results for all the locations show a marked decrease between 1981 and 1985/86.

Jensen and Bro-Rasmussen quote the results of German studies which again show a decline in cadmium-in-sediment concentrations over the past few years. The data in Table 21 are from their report.

Table 21

**Cadmium concentrations in sediments in German rivers  
(Jensen and Bro-Rasmussen, 1992)**

| River                   | Cadmium concentration in sediment, mg kg <sup>-1</sup> dw |      |      |
|-------------------------|---|------|------|
|                         | 1972  | 1979 | 1985 |
| Rhine, upper            | 5.3   | 3.1  | 2.1  |
| Rhine, lower and middle | 12.8  | 16.1 | 5.1  |
| Elbe                    | 17.0  | -    | 11.8 |
| Danube                  | 19.8  | -    | 2.1  |
| Weser                   | 13.6  | -    | 2.6  |
| Ems                     | 10.4  | -    | 1.7  |
| Main                    | 12.0  | -    | 3.9  |
| Neckar                  | 37.3  | 11.9 | 2.4  |

Jensen and Bro-Rasmussen also give some results from another study of sediments in the Rhine (Kramer and Duinker, 1986). The cadmium concentration (in the <16 mm fraction) near the centre of Rotterdam was about 30 mg kg<sup>-1</sup>, falling to about 1 mg kg<sup>-1</sup> at the mouth of the estuary; these values would be smaller if expressed in terms of total dry weight.

The above data suggest that concentrations of cadmium in river sediments of EU member states decreased by a significant amount during the 5 to 10 years preceding 1985. Few post-1985 data on cadmium-in-sediments have been found. However, cadmium concentrations in sediments are a reflection of cadmium concentrations in the rivers concerned. It may therefore be inferred from the water quality figures given in Table 19 that - for a number of rivers, including the Meuse - the improvement in sediment quality evident before 1985 has not been maintained. The elevated levels of cadmium entering the Netherlands via the Meuse during 1987/88 have already been noted (Table 6), and there can be no doubt that these levels gave rise to increased concentrations of cadmium in sediment.

Table 22

**Cadmium concentrations in different food items from various European countries  
(mg kg)**

| Food group or item              | Cadmium concentration, mg kg <sup>-1</sup> fresh weight <sup>a</sup> |                 |                 |                 |                  |                   |
|---------------------------------|--|-----------------|-----------------|-----------------|------------------|-------------------|
|                                 | NL <sup>b</sup>  | NL <sup>c</sup> | UK <sup>d</sup> | Dk <sup>e</sup> | Fin <sup>f</sup> | Swed <sup>g</sup> |
| Bread and cereals               | 18-34  | 25-30           | 21              | 30              | 20-40            | 31                |
| Meat                            | 2-3  | 10-40           | <10             | 6-30            | <5-5             | 2-3               |
| Offal                           |  |                 |                 |                 |                  |                   |
| pig kidney                      | 260  | 1000            | 450             | -               | 180              | 190               |
| pig liver                       | 44   | 100             | 130             | -               | 70               | 50                |
| Fish                            | 5  | 15              | <15             | 14              | <5-20            | 1-20              |
| Eggs                            | 1  | 2               | <30             | <10             | 1                | <10               |
| Oils and dairy products         | <30  | 10-30           | <5-30           | <30             | 3-20             | 1-23              |
| Sugars and preserves            | 5  | 5               | <10             | 30              | <10              | 3                 |
| Fresh fruit                     | 1  | 5               | <10             | 11              | <2               | 1-2               |
| Fresh vegetables (as a group)   | 15   | -               | <12             | -               | -                | -                 |
| Fresh vegetables (individually) |  |                 |                 |                 |                  |                   |
| cabbage                         | 5  | -               | <10             | 10              | 5                | 4                 |
| cauliflower                     | 5  | -               | <20             | -               | 10               | 10                |
| spinach                         | 60   | -               | -               | -               | 150              | 43                |
| broccoli                        | -  | -               | <20             | -               | 10               | -                 |
| legumes                         | -  | -               | <10-60          | 15              | <2-30            | 1-4               |
| lettuce                         | 30   | -               | <60             | 43              | 50               | 29                |
| potatoes                        | 30   | 30              | <30             | 30              | 30               | 16                |
| carrots                         | <40  | -               | <50             | -               | 30               | 41                |

## Notes

- (a) The concentrations are expressed as means unless otherwise stated. Where a range is given, this is the range of the means of sub-groups within a group - e.g. white bread, brown bread and other cereal products are three sub-groups within the "bread and cereals" group.
- (b) From Ros and Slooff, 1986; the reference cited is dated 1985.
- (c) From WHO, 1992a; the reference cited is dated 1988.
- (d) From MAFF (1983). The results are for the year 1981, except those for the individual vegetables - which are for 1976/77.
- (e) From WHO, 1992; the reference cited is dated 1979.
- (f) From WHO, 1992; the reference cited is dated 1980.
- (g) From WHO, 1992; the reference cited is dated 1984.

## 10. Concentrations in food

The cadmium content of foodstuffs in various countries has been reviewed by the WHO (1992). Additional data for the Netherlands and the UK are available from Ros and Slooff (1992) and MAFF (1983) respectively. A summary of some of these data is given in Table 22.

The two data sets for the Netherlands show some differences which are probably due to the difficulty of obtaining nationally representative samples. For example, it seems unlikely that there has been a tenfold increase in the cadmium content of meat in the interval between the two surveys. Indeed, Ros and Slooff (1988) note that during the period 1980-1985, the cadmium concentrations in pig liver and kidney decreased by about a factor of 2.

Van Assche and Ciarletta (1992) have noted a downward trend in the cadmium content of some general foodstuffs from Belgium and the Netherlands over the period 1979-1989.

The authors comment that, for foodstuffs derived from plants, the effect of decreasing atmospheric deposition is most evident those whose edible parts have been exposed to the atmosphere. Thus, for the "fruits" category, the cadmium content has fallen by a factor of about 3 in the decade considered. By contrast, the cadmium content of potatoes - a root crop - has fallen by a factor of only about 1.2. The sharp decline in the cadmium content of pig meat and kidney (by around a factor of 5) is possibly also due to a corresponding decrease in the cadmium content of their (plant-derived) feed. The decrease in the cadmium content of Dutch mussels reflects the general decrease in cadmium concentrations in the main river basins of Western Europe.

However, conclusions of this kind must be viewed with caution, especially when considering the very low concentrations found in most foodstuffs. The variations in analytical techniques over time, and differences in the methods used by different authors, make it difficult to draw firm conclusions about trends in dietary intakes of cadmium.

Crops grown in cadmium-contaminated areas have been shown to contain elevated levels of the metal compared with normal values. However, the bioavailability of soil cadmium depends on a number of factors - e.g. soil pH - and there is no universal relationship between soil cadmium and plant cadmium. Ros and Slooff (1988) report that in home-grown crops in the "Staadskanal" region, a 2, 3 and 7-fold increase in the cadmium content of potatoes, lettuce and endive respectively has been observed. In the Kempen, a 2-fold increase for potatoes, kale and sprouts, and a 7-fold increase for spinach and beetroot has been measured. The cadmium content of the total range of potatoes, vegetables and fruits in these areas is estimated to be, on average, a factor of 3 above normal values.

Table 23

Mean cadmium concentrations ( $\mu\text{g kg}^{-1}$ ) in selected crops grown at three contaminated sites in the UK, compared with normal values  
(adapted from WHO, 1992a)

| Location         | Source of cadmium contamination           | Cadmium concentration, $\text{mg kg}^{-1}$ fresh weight |             |        |        |
|------------------|---|---|-------------|--------|--------|
|                  |   | Cabbage   | Leafy salad | Potato | Carrot |
| Shipham          | zinc mine                                 | 250   | 680         | 130    | 340    |
| Walsall          | atmospheric inputs from a copper refinery | 73  | 190         | 103    | 120    |
| Heathrow         | application of sewage sludge              | 24  | 180         | 150    | 150    |
| Normal UK values |   | <10   | <60         | <30    | <50    |

WHO (1992) has tabulated cadmium concentrations in crops grown at three UK sites contaminated by three different kinds of cadmium source. Table 23 is adapted from these data.

Table 23 shows that the cadmium contents of the crops at the Walsall and Heathrow sites are a factor of about 3 or more higher than the normal UK values. The factors at Shipham range from about 4 to 25. These factors are about the same, or greater than, those reported for the Kempen. For the one vegetable, potato, common to both the Kempen and Shipham surveys, the cadmium concentrations for the Kempen and Shipham are about 60 and 130  $\mu\text{g kg}^{-1}$  respectively.

## 11. The mean daily intake from food

Although it is, of course, the cadmium contents of the individual food items which determine the average daily dietary intake of cadmium, this intake cannot be determined just from data of the kind given in Tables 20 and 21.

Three approaches may be used for estimating the daily intake of cadmium in food.

- The total diet study, in which the foodstuffs are prepared for consumption and then analysed individually or combined in groups in proportions based on the available food consumption data. The cadmium intake from any one food or food group is calculated as the product of the cadmium concentration and the estimated (national average) amount of that food eaten daily. The total

cadmium intake is found by summing the intakes for all the individual foods or foodgroups.

- The duplicate diet study, in which duplicate samples of all the meals consumed by the study group during the study period are collected. The combined food sample for each individual is homogenised and analysed for cadmium.
- The daily faecal output study. Faecal analysis may be used to determine the cadmium intake because only about 5% of ingested cadmium is absorbed.

Some reported estimates from various countries of the mean daily intake of cadmium in food are given in Table 24.

Where both a total diet study and a duplicate diet study are carried out on the same group, the duplicate diet study usually gives the lower results. The reasons for this are not clear; the effect is evident in Table 23. There is also, in Table 24, some suggestion of a decline in cadmium intakes during the 1980s.

The examples given in Table 24 of mean daily intakes estimated for people living in contaminated areas and eating home-grown crops refer to group means. Clearly, there will be individuals within the group who have higher than average cadmium intakes. For example, in the duplicate diet study carried out in Shipham, UK, the average daily intake of the exposed group was 26  $\mu\text{g}$ , but the highest individual daily intake was 150  $\mu\text{g}$  (Morgan, 1988).

It is clear from Table 24 that - if like is compared with like, as regards e.g. type and year of study, and type of area - the mean daily intake of cadmium in food in most EU member states is relatively similar.

## 12. The mean daily intake from all sources

Cadmium may be taken into the body not only via food, but also via drinking water, the inhalation of the cadmium normally present in the atmosphere, and the inhalation of cigarette smoke.

Drinking water is usually a minor contributor to the total cadmium intake for the general population. The WHO in their Guidelines for drinking-water quality (WHO, 1993) note that levels in drinking-water are usually less than 1  $\mu\text{g L}^{-1}$ , and that food is the main source of daily exposure to cadmium. If a daily per capita consumption of 2 L is assumed, then a concentration in drinking water of 1  $\mu\text{g L}^{-1}$  would give rise to a daily intake of cadmium of 2  $\mu\text{g}$ .

Table 24

Estimates from various countries of the mean daily intake of cadmium from food

| Country                              | Type of study <sup>a</sup> | Year of study | Cd intake, $\mu\text{g d}^{-1}$ | Reference                      |
|--------------------------------------|----------------------------|---------------|---------------------------------|--------------------------------|
| The Netherlands                      | TD                         | 1985          | 20                              | Ros and Slooff, 1988           |
| The Netherlands                      | DD                         | 1985          | 9                               | Van Assche and Ciarletta, 1992 |
| The Netherlands (contaminated areas) | TD                         | 1985          | 35 <sup>b</sup>                 | Ros and Slooff, 1988           |
| UK                                   | TD                         | 1988          | 11-18 <sup>c</sup>              | OECD, 1994                     |
| UK (contaminated area)               | TD                         | 1979          | 36 <sup>d</sup>                 | WHO, 1992                      |
| UK (contaminated area)               | DD                         | 1979          | 26 <sup>d</sup>                 | Morgan, 1988                   |
| Germany                              | DD                         | 1986/87       | 10                              | OECD, 1994                     |
| Germany (Bavaria)                    | TD                         | 1989          | 13                              | OECD, 1994                     |
| Belgium                              | TD                         | 1980          | 45                              | Van Assche and Ciarletta, 1992 |
| Belgium                              | DD                         | 1981          | 18                              | Van Assche and Ciarletta, 1992 |
| Denmark                              | TD                         | 1980          | 32                              | Morgan and Sherlock, 1984      |
| Denmark                              | TD                         | 1983/87       | 20                              | OECD, 1994                     |
| France                               | TD                         | 1976/78       | 29                              | Morgan and Sherlock, 1984      |
| France                               | DD                         | 1983/90       | 19                              | OECD, 1994                     |

## Notes

- (a) TD = total diet; DD = duplicate diet
- (b) For persons living in the Stadtskanal and Kempen regions, who consume home-grown crops. In arriving at their estimate of 35 mg per day, Ros and Slooff assume that all the potatoes and other vegetables eaten by the group are home-grown. This is unlikely to be true for the whole year, and the estimate is therefore probably too high.
- (c) The lower figure assumes concentrations below the limit of measurement are zero, the higher figure assumes that they are equal to the limit of measurement.
- (d) For persons living in Shipham, who consume some home-grown crops.

Some measured values of cadmium in drinking water in the UK are given in the report of the national total diet study (MAFF, 1983). Of the 89 samples taken, 84 had concentrations no greater than  $2 \mu\text{g L}^{-1}$ , and only one had a concentration in excess of  $4 \mu\text{g L}^{-1}$ . For the Netherlands, Ros and Slooff (1988) report the results of a study of drinking water quality in 1982: in about 99 per cent of the samples taken from pumping stations, and in 256 peripheral samples, the concentration was less than  $0.1 \mu\text{g L}^{-1}$ . A concentration of  $0.1 \mu\text{g L}^{-1}$  would give rise to a daily intake of cadmium of only  $0.2 \mu\text{g}$ . Such an intake is trivial compared with the  $9 \mu\text{g d}^{-1}$  (duplicate diet study) or  $20 \mu\text{g d}^{-1}$  (total diet study) taken in via food.

The average concentration of cadmium in air, in urban areas of European countries is now probably no more than  $3 \text{ ng m}^{-3}$  (see Table 5). Assuming a daily (24h) inhalation rate of  $15 \text{ m}^3$ , an absorbed fraction for inhalation of 25 per cent and an absorbed fraction for ingestion of 5 per cent (WHO, 1992a), an airborne concentration of  $3 \text{ ng m}^{-3}$  would lead to an equivalent oral intake of about  $0.2 \mu\text{g}$ . Calculations of this kind assume that the concentration of cadmium in indoor air is about the same as in outdoor air - the measurements given in Table 8 refer to outdoor air, but most people spend most of their time indoors. Ros and Slooff refer to studies in the US which indicate that indoor and outdoor concentrations are usually much the same, except in houses where smoking takes place - where the cadmium concentration may be around  $100 \text{ ng m}^{-3}$ . A person exposed to this level for several (say, 5) hours per day would receive an equivalent oral intake of about  $1.5 \mu\text{g}$ .

Smokers themselves will of course receive cadmium intakes both directly from the inhalation of mainstream smoke, and indirectly from passive smoking. Smoking a pack of 20 cigarettes daily can result in the direct inhalation of 2-4 mg cadmium (WHO, 1992a), and this is equivalent to an oral intake of about 15 mg. The equivalent oral intake from, say, 10 hours passive smoking would be about 3 mg, leading to a total equivalent oral intake from smoking of about 18 mg per day. This is about the same, or greater than (depending on whether the total diet or duplicate diet figure is taken), the intake via food.

It may be concluded that the mean daily per capita intake of cadmium (expressed in terms of an equivalent oral intake) for the Dutch population - averaged over the year and averaged over the whole population is about 20 mg for non-smokers and 40 mg for smokers (one pack per day). These figures are based on the total diet studies for determining the intake via food.

There will, of course, be individual members of the population whose mean daily intake of cadmium is very different from the population mean. Heavy smokers fall into this category, as well those living in contaminated areas and eating home-grown produce.

### 13. The tolerable daily intake

The derivation of a tolerable long-term cadmium intake has been discussed by the Joint FAO/WHO Expert Committee on Food Additives - JECFA (WHO, 1989).

The kidney, in particular the renal cortex, has been identified as the target organ, and the first functional change is usually an increased excretion of low molecular weight proteins. This is not accompanied by any adverse histological changes and its pathological significance is unclear. Further accumulation of cadmium may cause disturbances in the reabsorption of amino acids, glucose and minerals. The only way to avoid chronic renal dysfunction is to prevent cadmium concentrations in the renal cortex reaching a critical value of around 200 mg kg<sup>-1</sup> wet weight. This is not a no-effect-level, but rather the critical concentration for a small fraction (around 10 per cent according to JECFA) of the exposed population - i.e. the critical concentration for the most sensitive group. If assumptions are made as to the absorption of cadmium from the gut, and as to the excretion rate of the cadmium body content, then it is possible to estimate the concentration in the renal cortex produced by a given daily dietary intake over a given number of years. The most recent estimations made by JECFA (WHO, 1989) are as follows: assuming an absorption rate for dietary cadmium of 5 per cent, and a daily excretion rate of 0.005 per cent of body content, it may be concluded that the average daily dietary intake should not exceed about 1 mg per kg of body weight per day continuously for 50 years if levels of cadmium are not to exceed 50 mg kg<sup>-1</sup> in the renal cortex. JECFA chose this level (a factor of 4 below the critical level of 200 mg kg<sup>-1</sup>) as an acceptable one, and therefore set their Provisional Tolerable Weekly Intake (PTWI) at 7 mg per kg body weight. Taking the body weight of an adult to be 70 kg, this PTWI corresponds to a tolerable daily intake of 70 mg.

The mean daily intake for non-smoking populations in Europe who do not eat home-grown produce in contaminated areas, is about 20 mg - much less than the tolerable daily intake. UK data (unpublished) indicate a 97.5 percentile daily intake value of 30 mg for the general population. For the small number of individuals who eat home-grown produce in contaminated areas, and who smoke a pack of cigarettes a day, the mean daily intake might be expected to be about 35 (Table 23) plus 18 mg, say 55 mg. This is still less than the tolerable daily intake. However, as was mentioned earlier, it is almost certain that there will be some individuals - at the top end of the most highly exposed groups - whose mean daily intake does exceed the tolerable daily intake. With the data currently available, it is not possible to quantify the numbers of such individuals or the extent of their exposure.