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The relative contribution of different environmental sources to human cadmium exposure and the EU cadmium risk assessment

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Abstract

A stepwise model is presented, in which the stages of the relevant pathways of cadmium transfer from the environment to man are identified and quantified. Normalisation of the data at each of the subsequent stages of the pathways results in a quantitative assessment of the relative importance of the different environmental sources that are ultimately at the origin of the exposure of the general population to cadmium.

The model is applied to averaged recent data for the EU, showing the significant contribution of natural cadmium to current human exposure. P-fertiliser application to agricultural soils and industrial sources of cadmium emission to the atmosphere are identified as most relevant man-made inputs. The production and use of cadmium containing products does not contribute at all to human exposure.

The model allows for the introduction of data characteristic for local exposure situations.

1. Introduction

Cadmium has been under strong regulatory attention since the 1970s. As a result of this, environmental levels of cadmium, its sources and pathways to human exposure are well known and documented since several decades. This monitoring demonstrates a clear and general downward trend in anthropogenic cadmium emissions (a.o. *Jenssen and Bro-Rasmussen 1990, OECD 1993, Hutton 1995*) and human exposure (*Van Assche and Ciarletta 1993*). This resulted in the early 1990s in a daily cadmium exposure of the general population « at the lower end of the range 10-25 µg » (IPCS 1992). This actual exposure can be compared with the provisional tolerable daily intake value of 60-70 µg/day, set by the WHO.

Recently, there is renewed regulatory attention in the EU, mainly on Cd-products. The use of cadmium in pigments, in stabilisers and as a plating agent is subject of a risk assessment for human health and environmental effects within the framework of the Accession treaty discussions, as well as the revision of directive 91/338 on the restrictions of marketing and use of cadmium in pigments, stabilisers and plating. The battery directive 91/157, which includes NiCd batteries, is also under revision, and NiCd batteries have received specific regulatory attention in the Northsea Conference programme.

Within the framework of the EU « Existing Substances » Regulation 93/793, cadmium metal and cadmium oxide were put on the 3rd list of priority substances (January 1997) for which a EU risk assessment should be made. In this regulatory process, the Cd/CdO-producing industry is legally obliged to provide the EU competent authority (the « Rapporteur »; in the case of Cd/CdO: Belgium) with an extensive file containing all relevant information on the toxicology and ecotoxicology of these priority substances and, most importantly, on human (general and occupational) and environmental (general and production site-specific) exposure. The exercise encompasses the whole life cycle of the substances, including their use in products, recycling and/or disposal. In the case of Cd/CdO, focus is put on NiCd batteries, given the fact that the other applications of the two substances (in pigments, stabilisers and plating) are assessed in the specific risk assessment mentioned above. The EU risk assessment on Cd/CdO is ongoing and is making good progress; the report of the Belgian Rapporteur is expected during the first half of 1999.
With the exception of the latter EU risk assessment, most regulatory discussions on Cd are focused on a particular aspect, i.e. on Cd-products, Cd in fertilisers, There are, however, many sources of Cd in the environment (including natural), which contribute all to human exposure. Quantitative assessments of the proportional contribution of all different Cd sources to human exposure are scarce.

In this paper, an attempt to such integrated approach is made. The analysis is based on a) well-established quantitative knowledge on the main transfer pathways of cadmium from the environment to the general population, and b) recent monitoring and emissions data.

2. Methodological approach

In a stepwise model, the relevant stages of the main pathways of Cd-transfer from the environment to man are identified and quantified in a normalised way. The stepwise model is presented in *Figure 1*. By normalising at each of the subsequent stages, a quantitative assessment can be made of the relative importance of all the different cadmium sources that are ultimately at the origin of human exposure.

The analysis presented in this paper is related to the general, non-smoking population, living in an environment not contaminated by direct point source emissions. Data at the different stages of cadmium transfer are averaged. Emissions are mostly based on an inventory for the EU, made at the end of the 1980s (ERL 1990); some updates, taking into account recent data, are included. As such, a general quantitative analysis is made, which is representative for the EU in the 1990s.
It is clear that locally; the contribution of different parts of the transfer pathways can vary considerably. The model allows for such specific entries, reflecting local conditions. The relative importance of such variations will be discussed.

It should be emphasised that the current analysis is based on total cadmium mass transfer. The solubility and hence the bioavailability (availability for uptake by organisms) of the Cd-forms released will determine their potential for transfer to man. This bioavailability can be quite different for the physico-chemical forms released by different sources. Moreover, cadmium is a natural constituent of the earth's crust and as such present in the environment, including food, in a natural way. An assessment of this natural component of human Cd-exposure is also included in the present analysis.

3. Entries and assumptions of the model

(Read figure 1 from top to bottom**)**

Step 1: 100% internal Cd-exposure is the starting point, defined by two routes of internal cadmium: oral ingestion and inhalation.

Step 2: main routes of human exposure are: a) oral ingestion, b) inhalation of ambient air (non-smokers(*)). The ratio oral ingestion / inhalation: 96% / 4%, based on: a) a daily intake of 15 µg (IPCS 1992), a gastro-intestinal absorption: 5%, resulting in an internal dose of 0,75 µg/day, and b) inhalation of 12 m^3 of ambient air/day with a concentration of 5 ng/m^3(*) (urban; Hutton 1995), and a lung absorption of 50%, resulting in 0,03 µg/d

Step 3: a) sources of Cd in daily diet are:

- Drinking water: 1% of total (average concentration in drinking water < 0,1 µg/l, resulting in (2 l consumption/day) < 0,2 µg/d intake. Compare to 15 µg from food (Hutton 1995)
- Food from terrestrial origin (plants, animals): 98% and food from aquatic origin (fish, seafood,): 1% (Fouassin & Fondu 1981, CCRX 1991)

b) Sources of Cd in ambient air: see « sources of Cd to the atmosphere »

Step 3bis: crops contain Cd due to a) root uptake and translocation to edible plant parts, and b) direct atmospheric deposition of Cd on edible plant parts. The ratio between a) and b) is considered to be 70/30 (Hovmand et al 1983, Harrison & Chirkawi 1989). The Cd-content of animal food is considered being also of vegetative origin. The main part of this Cd is excreted (95%) in manure, which can be returned to soils as a fertiliser (Landner et al 1996). This « recycling » of Cd to soils is therefore not considered in stage 4 (sources of Cd to soil), and the contribution of plant and animal food is taken together as « terrestrial » food.

(*) Hutton (1995) notes that in houses where smoking takes place, the cadmium concentration in air may mount to 100 ng/m^3. A person exposed to this level for several (e.g. 8) hours would receive an internal dose of about 0,150 µg/day. Smoking a pack of 20 cigarettes a day results in an internal dose of 20 x 1,5 µg Cd/cigarette x 10% inhaled x 50% absorbed = 1,5 µg/day.

Step 3 Terri: Apart from Cd added to soil by human activity, soils contain natural Cd which will result in background Cd-levels in plants.

Natural and ambient Cd levels in soil are dependent on soil type and location and vary considerably within the EU. For natural levels, an average value of 0,1 µg/g is generally cited (Ros & Slooff 1990, IPCS 1992); for “normal” agricultural soils in the EU, an average of 0,44 µg Cd/g (0,12-1,0 µg/g) is reported (Hutton 1995; non-contaminated areas). Accordingly, the natural Cd in soils has been estimated for this analysis to contribute at least 20 % to total plant Cd uptake. It should be emphasised, however, that the soils used to make this estimation are not the same, and this comparison should therefore be made with great care.

Indeed, in a retrospective analysis, going back to the mid 1800s, Jones et al (1987) demonstrated that the total atmospheric (considered to be entirely antropogenic) Cd-input on experimental soils over that period has resulted in an increase of the soil Cd-level of about 27-55 %. The additional input from (low) P-fertiliser applied to adjacent plots was roughly estimated as similar. From these data it could be estimated that since the industrial revolution, the Cd content of soils might have doubled due to antropogenic influence, suggesting a 50/50 ratio for the natural/antropogenic Cd content of agricultural soils.

On the other hand, the antropogenic emissions to air have been significantly higher in former times, e.g. during the 1960s, than at present. Consequently, there is no direct relationship between the atmospheric emission pattern of the early 1990s (table 1) and the antropogenic inputs through atmospheric deposition since the previous century. The ratio between the natural and antropogenic soil Cd, used in the present analysis (20/80) should therefore be considered with great caution and most probably represents a worst case generalisation. It is clear that the ratio is strongly influencing the end result of the analysis, taken into account the relative importance of the soil-plant-food transfer pathway for human exposure.

Step 4: antropogenic sources of Cd to soils are: P-fertiliser application, Atmospheric deposition and application of sewage sludge in a 50/30/10 ratio (based on Jensen & Bro-Rasmussen 1992, Stigliani and Jaffe 1993, Landner et al 1996). P-fertiliser is rather generally spread over large agricultural areas. Local differences can occur due to differences in application rate and Cd-content of the fertiliser used. Application of sewage sludge and farmyard manure differs strongly between countries (Landner et al 1996). The « recycling » of Cd through farmyard manure was not included as an additional Cd source to soils.

Step 5: a) Cd sources to air have a direct impact on inhaled Cd, and after deposition on : Cd in soil (30% of total of soils), Cd in exposed plant parts (30% in total in plants), Cd in surface waters (15% of total emissions is deposited to surface waters).

b) Cd sources to surface water contribute to Cd-content in sludges, and thus in soil, and directly to the Cd-content of food from aquatic origin. If surface waters (instead of groundwater) are used for drinking water, there is direct influence on this pathway also; in this analysis, drinking water is assumed to originate entirely from surface waters. Treatment of waste waters effluents and surface waters results in sludges that can be applied to agricultural soils. Inn this analysis, it is assumed that all sludges are applied to agricultural soils. It is further assumed that 50% of surface water and effluents is treated; as a result of treatment, 70% of the Cd input is found in the sludge, 30% remains in the surface water fraction. Therefore, 35% of the Cd from sources to water is going to sludge, 65% remains in the water. It should be noted that industrial waste waters are usually treated separately on site, and the resulting sludges are stored in controlled landfill, surely not used for application on agricultural soils. The Cd in sludges thus has different origins, including natural erosion, household emissions, minor industrial activities, atmospheric deposition. For this analysis, it is however assumed that the sources as listed in *Table 1* contribute proportionally to the Cd in sludge.

The emissions of different sources of Cd to air and water in the EU are listed in *Table 1*. This table is based on an extensive survey for the EU made at the end of the 1980s (ERL 1990). However, for some sources, the downward trends in Cd emissions have continued since then. Therefore, some of the ERL data are updated with more recent information:

- Surface waters have a natural background Cd level of about 70 ng/l (Ros & Slooff 1990). The average Cd level of EU rivers during the 1980s was 350 +/- 300 ng/l (Hutton 1995). The contribution of natural Cd in surface waters is thus estimated at 20%.

- The releases to water from P-fertiliser manufacture were revised downward including more recent information from the Netherlands (Coppoolse et al 1992) and Belgium (Technical Commission Northsea 1995).

- The atmospheric deposition to surface water was estimated at 15% of total atmospheric release, based on data from the Netherlands (Coppoolse et al 1992).

- The release to water from the zinc industry was revised downward to 1 tonne, taking into account recently reported emissions for 1996 from EU producers (average emission factor to water: 175 g Cd /t of Cd produced).

4. Calculations

a) Sources to the atmosphere

(Deposition on soil: 0.3 x 0.8 x 0.7 x 0.98 x 0.95 = 15.81%) + (deposition on plants: 0.3 x 0.98 x 0.96 = 28.22%) + (direct inhalation = 4%) + (sludge to soil: 0.15 x 0.5 x

0.7 x 0.1 x 0.8 x 0.7 x 0.98 x 0.95 = 0.3%) + (deposition to water: (cf. *Table 2*) = 0.55%) = **48.9%**

b) Sources to the water

(sludge to soil: 0.5 x 0.7 x 0.1 x 0.8 x 0.7 x 0.98 x 0.95 = 1.84%) + (direct to aquatic food : 0.65 x 0.01 x 0.96 = 0.624%) + (direct to drinking water : 0.65 x 0.01 x 0.96 = 0.624%) = **3.1%**

c) P-fertiliser direct to soil

0.6 x 0.8 x 0.7 x 0.98 x 0.96 = **31.61%** (+ P-fertiliser to water term)

d) Natural Cd

to soil: 0.2 x 0.7 x 0.98 x 0.96 = **13.2%** + natural to air term + natural to water term

e) The relative contribution of the different sources of Cd to air and water in a perspective of human exposure can be calculated from *Table 1*, taking into account 48.9% contribution from atmospheric sources to total exposure, and a total contribution of 3.1% from water sources to total exposure: *Table 2*.

Combination of the information from a) → e) results in the following end result (see also *Figure 2*):

- natural sources : 20.22% (13.2% to soil + 6.4 % to air + 0.62 % to water)
- P-fertiliser: 32.45% (application to soil: 31.61% + production to water: 0.84%)
- Fossil fuel combustion: 21.5% (to air)
- iron and steel production: 11.2% (to air : 10.5% + to water : 0.7%)
- non-ferrous metals production: 6.3% (5.9% to air + 0.4% to water)
- production and use of Cd products: 0.54% (0.5% to air + 0.04% to water)
- cement manufacture : 3.1% (to air)
- waste incineration : 1.1% (to air)

5. Discussion

The main general conclusions from a perspective of human exposure to Cd following from the present analysis are:

- inputs of Cd to soil are most important, due to the dominance of ingestion of food from terrestrial origin for human exposure. For this reason, application of P-fertiliser is quantitatively important.

- emissions to atmosphere are quantitatively more important than emissions to water. The latter category is in fact rather insignificant for human exposure.

- the natural contribution of Cd to human exposure is significant (1/5 of total, taking into account a 20/80 ratio between natural and antropogenic soil Cd. This ratio may, as indicated, be more close to 50/50, which would increase significantly the relative contribution of natural Cd to human exposure).

- the production and use of Cd products (pigments, stabilisers, plating, alloys, NiCd batteries) does not contribute at all to human exposure.

The analysis applies to the non-smoking general population. Smoking would add about 1.5 µg to the daily dose, or would, in other words, more than double the daily Cd dose.

The quantitative assessment of the relative contribution of different emission sources presented in this paper depends on the quantitative emissions data that are used. As mentioned, the data inputs are averaged for the situation in the EU in the early 1990s. It is clear that any change in emission pattern related to the sources listed will influence the quantitative result. Indications of such changes (e.g. significant decreases of emissions) have been observed for e.g. the combustion of coal to air and P-fertiliser production (T.C.N. 1996).

Considering the major pathways of Cd for human exposure, the variation that can most influence human exposure would be smoking habits, different input through P-fertilisers, vicinity to heavy atmospheric point source.

Smaller changes in exposure could result from different dietary habits (e.g. strong consumption of seafood), and vicinity of moderate atmospheric emission sources.
Populations living in the immediate surroundings of former point sources e.g. smelters can have higher exposure due to consumption of contaminated groundwater as drinking water, and through enhanced soil-plant Cd transfer (soils with low Cd-complexing capacity). The quantitative modelling presented in *Figure 1* allows integrating local data for the respective parameters.

The contribution to human exposure from landfill is considered minimal in this analysis. Industrial waste deposits containing Cd are nowadays properly designed and controlled to prevent leakage of Cd to underlying groundwater and surface water. Leakage has occurred at older industrial dumps; these are being controlled progressively. Municipal waste dumps contain Cd from natural origin (e.g. plant material) and from discarded Cd products. In most products, Cd is firmly contained in a matrix (e.g. pigments, stabilisers) and therefore has very limited mobility. The release of Cd from landfilled NiCd batteries is also very limited (Oda 1995). As a matter of fact, the Cd release from landfills was observed to be very low indeed (Eggenberger and Waber 1997).
Finally, it has to be emphasised again that the present analysis is based entirely on mass transfer. Differences in the bioavailability between the Cd forms emitted to the environment and applied to soil will influence the results of the quantitative assessment to some extent, too.

Table 1: Sources of Cd to air and water in the EU
Data based on ERL (1991). Updated figures (industry emission figures) are indicated in italics

	To air (t/a)	**%**	**To water (t/a)**	**%**
a) Natural sources	14,7	13,1	*18,93*(1)	20
b) antropogenic sources • combustion of fossil fuels(2)	49,4	43,9	n.s.	
• iron and steel production	24,2	21,5	21,2	22,4
• non-ferrous metals production(3)	*13,6*	12,1	*11,0*	11,6
• production and use of Cd products(4)	*1,04*	0,93	*1,13*	1,2
• production of P-fertilisers	n.s.	n.s.	*25,7*	27,1
• cement manufacture	7,0	6,2	n.s.	
• waste incineration(5)	*2,5*	2,2	n.s.	
• atmospheric deposition(6)	-	-	*16,7*	17,6
	112,44	**100**	**94,7**	**100**

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(1) 20% of total.

(2) In Belgium, a strong decrease of this source was observed over the period 1985-1995 (Technical Commission Northsea, 1996).

(3) Updated based on 1995-1996 data.

(4) Figures from ERL were updated as follows : emissions (t/a) from :
- NiCd battery manufacturing : 0,23 to air - 0,16 to water (OECD 1995)
- Cd pigment manufacturing : 0,012 to air - 0,025 to water (WS Atkins 1998)
- Manufacturing and use of Cd stabilizers : 0,002 to air - 0,005 to water (WS Atkins 1998)
- Cd plating : n.s. to air - 0,940 to water (WS Atkins 1998)
- Cd alloys : not updated.

(5) ERL (1990) estimated the Cd-capturing capacity of waste incinerators in the EU to be 88%. More recently, modern technology can mitigate releases of Cd to air by >99%. Directives and standards have been installed to reach this efficiency e.g. the waste incineration directives 89/369 and 89/429 that set emission limit values to air based on BAT for respectively new and existing municipal waster incinerators. Recently, incinerators in most EU countries have reached such high efficiency except for some countries. Considering that now also such incinerators have been progressively closed down, the efficiency of Cd retention of EU incinerators is set at 99%.

(6) Contribution of atmospheric deposition to surface water : 25% of total in Netherlands (Coppoolse 1992). Therefore assumed to average 15% for EU.

n.s. : considered not significant.

Table 2: Relative contribution (%) of Cd sources to air and water in a perspective of total human exposure

	To air	To water
- Natural sources	6.4	0.62
- Combustion of fossil fuels	21.5	n.s.
- Iron and steel production	10.5	0.7
- Non-ferrous metals production	5.9	0.4
- Production and use of Cd products	0.5	0.04
- Production of P-fertilisers	n.s.	0.84
- Cement manufacture	3.1	n.s.
- Waste incineration	1.1	n.s.
- Atmospheric deposition	-	0.55

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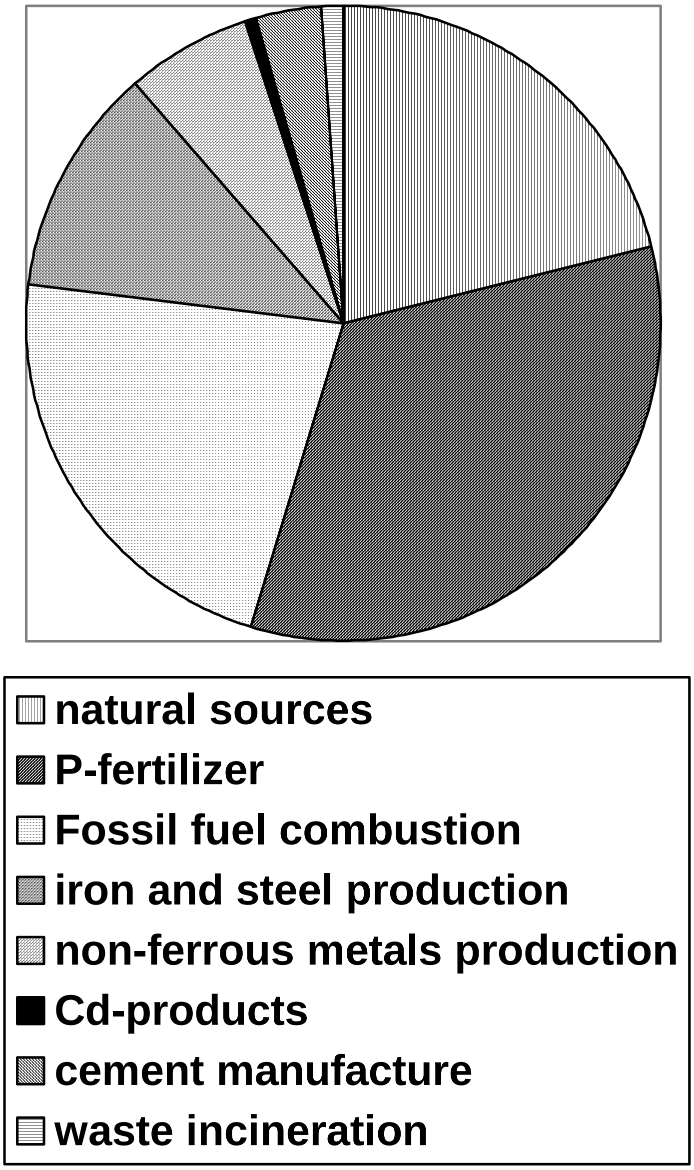
Figure 1: Pathways of human exposure to Cadmium
See powerpoint file page attached

Figure 2:



Relative contribution (%) of the different sources of cadmium to human exposure