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The Relative Contributions of Different Environmental Sources to Human Exposure and the EU Cadmium Risk Assessment

-an update of figures to the presentation at the NiCd Conference in Prague, 1998 (Van Assche, 1998)

By Lidia Regoli.
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Abstract

At the NiCd 98 Conference in Prague, a stepwise model was presented, in which the stages of the relevant pathways of cadmium transfer from the environment to man were identified and quantified. Normalisation of the data at each of the subsequent stages of the pathways resulted 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.
In this paper, the model is once again applied, using updated data from the Cd risk assessment for the EU. The conclusions are the same as 7 years ago. The model shows 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. Furthermore, the model allows for the introduction of data characteristic for local exposure situations.

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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 general downward trend in anthropogenic cadmium emissions (Boutron et al, 1995, North Sea Conference, 1995 and Staessen et al.,1999) and human exposure (Van Assche and Ciarletta, 1993). The WHO has estimated a daily cadmium exposure of the general population in the range of 24 to 42 µg (JEFCA, 2003), however levels are generally lower in European countries (10-35 µg) (EU Cd RA, 2004). This actual exposure can be compared with the provisional tolerable daily intake value of 60-70 µg/day, set by the WHO (JEFCA, 2003).

Recently, there is renewed regulatory attention in the EU, mainly on the revision of the EU battery directive 91/157 including within its scope the Ni-Cd batteries. In fact, NiCd batteries have received specific regulatory attention within the revision of this directive, and within the North Sea Conference Programme.

Within the framework of the EU "Existing Substance" Regulation 93/793, cadmium metal and cadmium oxide were put on the 3rd list of priority substances (January 1997) for which conducting a EU risk assessment was required. In this regulatory process, the Cd/CdO-producing industry was 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 encompassed the whole life cycle of the substances, including their use in products, recycling and/or disposal. In the case of Cd/CdO, focus was put on NiCd batteries, given the fact that the other applications of the two substances (in pigments, stabilisers and plating) were assessed in the specific risk assessment for human health and environment within the framework of the revision of the directive 91/338 on the restrictions of marketing and use of cadmium pigments, stabilisers and plating. The EU risk assessment on Cd/CdO has now been finalised for human health, and is in the final stages for environment and for the targeted risk assessment on NiCd batteries. For the latter two, final draft reports have been submitted for final comments.

In the meantime, a risk reduction strategy is underway, in which the Belgian Rapporteur has the mandate to make recommendations for a risk reduction strategy based on the conclusions of the risk assessments.
Within this context, the relative contribution of sources of cadmium to human exposure becomes important in order to understand the relative impacts of different reduction strategies.

With the exception of the latter EU risk assessments, most regulatory discussions on Cd are focused on a particular aspect, e.g. 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 cadmium sources to human exposure have been highlighted by F Van Assche at the Ni-Cd Conference in Prague in 1998. This analysis was based on :
a) well established quantitative knowledge on the main transfer pathways of cadmium from the environment to the general population,
b) recent monitoring and emissions data.

In this paper, the approach taken by Van Assche 1998 (described in detail in the annex) is revised with more recent data available from the EU Cd risk assessment.

2. methodological approach

The stepwise model with the relevant stages of the main pathways of Cd-transfer from the environment to man is described in detail in Van Assche 1998 (entries and assumption of the model are described in the annex), and summarized in Figure 1. A quantitative assessment has been made of the relative importance of all the different cadmium sources that are ultimately leading to human exposure.

The analysis summarized in this paper is related to :

1) the general, non-smoking population, living in an environment not contaminated by direct point source emissions
2) the general smoking population living in an environment not contaminated by direct point source emissions.

Data at the different steps of cadmium transfer are averaged. Emissions are mostly based on an inventory for the EU, made at the end of the 1980s (ERL, 1990), with some recent updates taking into account more recent changes in industry production figures and EU monitoring databases. As such, a general quantitative analysis is made, which is representative for the EU in the late 1990s, early 2000s.
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 also 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 also determine their potential for transfer to man. This bioavailability can be quite different for the physico-chemical forms of Cd 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. Discussion

A summary of the tonnages per year and relative contribution of different sources of cadmium to air and water as presented in the EU Cd risk assessment is summarized in Table 1. This also includes the contribution from natural sources. This information

was used as data input for assessing the relative contribution of Cd sources to air and water in a perspective of total human exposure. The summary of the evaluation is described in Table 2.

Table 1 : Sources of Cd to air and water in the EU

Data based on the Cd EU risk assessment (compiled from ERL (1990) figures and updated figures (industry emission figures, recent EU monitoring databases)

	To air (T:yr)	%	To water (T/yr)	%
a) Natural sources	15	11	23,6[1]	30
b) Anthropogenic sources				
- combustion of fossil fuels[2]	55,7	41	0,1	0,1
- iron and steel production	31	23	15,6	21,9
- non-ferrous metals production[3]	11	8	9,9	13,9
- production and use of Cd products[4]	1,2	1	0,3	0,4
- production of P-fertilisers	0,7	0,5	9,1	12,8
- Municipal waste incineration	3,2	2	1,6	2,2
- other (cement manufacture, traffic,..)	19	14	1,7	2,4
- atmospheric deposition[6]	--	--	16,7	23,4
	136,8	**100**	**78,6**	**100**

(1) 30% is considered natural Cd (FOREGS 2004 compared with average 90P of monitoring databases for 7 countries)
(2) In Belgium, a strong of this source was observed over the period 1985-1995 (Technical Commission Northsea, 1996)
(3) Update of Cd production figures from 2003 (zinc figures date to 1996)
(4) WS Atkins (1998) and data from TRAR (Draft version of May 2003)
(5) Estimated Cd-capturing capacity of wase incinerators in the EU >99% (OECD, 1996)
Directives and standards have been installed to reach this efficiency
(waste incineration directives 89/369 and 89/429 setting emission limit values based on BAT).
(6) Contribution of atmospheric deposition to surface water : 25% of total in NL (Coppoolse, 1992).

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

	To air	To water
Natural sources	5,3	1
Combustion of fossil fuels	19,6	n.s.
Iron and steel production	11	0,7
Non-ferrous metals production	3,8	0,4
Production and use of Cd products	0,3	n.s.
Production of P-fertilisers	n.s.	0,4
Municipal waste incineration	1	n.s.
Other (cement manufacture, traffic,..)	6,7	n.s.
Atmospheric deposition	--	0,75

n.s. <0,1%

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 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 anthropogenic 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 population. Smoking adds about 1,5 µg to the daily dose, or would, in other words, 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 1990s.

It is clear that any change in emission pattern related to the sources listed will influence the quantitative result. Indications of such changes (significant decreases of emissions) have been observed in Belgium for the emissions to air following from the combustion of coal and for the emissions to water from 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:
1) smoking habits
2) different input through P-fertiliser
3) vicinity to heavy atmospheric point source

Smaller changes in exposure could result from different dietary habits (e.g. strong consumption of seafood), vicinity of moderate atmospheric emission sources. Populations living in the immediate surroundings of former atmospheric 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 to integrate 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 in order 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, personal communication). As a matter of fact, the Cd release from landfills was observed to be very low (Eggenberger and Waber, 1997). Furthermore, it should be noted that effective collection and recycling schemes have been established across the EU for NiCd batteries.
Finally, it has to be emphasised again that the present analysis is based entirely on mass transfer. Differences in bioavailability between the cadmium forms emitted to the environment and applied to soil will influence the results of the quantitative assessment to some extend, too.

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ANNEX

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 is 98%/2%°, based on a daily intake of 30 µg (JEFCA, 2003)[1], a gastrointestinal absorption of 5%, resulting in an internal dose of 0,75 µg/day. Inhalation of 12 m^3 ambient air/day with a Cd concentration of 5 ng/m^3 (urban, Hutton 1995, target limit value for Cd in the Air Quality Directive), and a lung absorption of 50%, results in 0,03 µg/d

Step 3 :

a) sources of Cd in daily diet are :
   - drinking water : 1% of total (average Cd concentration in drinking water : <0,1 µg/l, resulting in (2 l consumption by day) < 0,2 µg/d intake. Compare with 30 µg from food (JEFCA, 2003 and 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 part, and
b) direct atmospheric deposition of Cd on edible plant parts. The ratio between a) and b) is considered 70/30 (Hovmand et al. 1983, Harrison & Chirkawi, 1989). The Cd-content of the 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".

Step3ter : 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 and 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 (non-contaminated areas) by Hutton (1995). Accordingly, the natural Cd in soils has been roughly estimated for this analysis to contribute at least 20% to total plant Cd uptake. It

[1] This is a revised figure from what was presented by F Van Assche in 1998, based on JEFCA 2003 and represent the upper range for Europe (according to data compiled in the Cd EU risk assessment).

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 caution.
Indeed, in a rare retrospective analysis, going back to the mid-1800s, Jones et al (1987) demonstrated that the total atmospheric (considered to be entirely anthropogenic) Cd-input on experimental soils over that period had 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 being similar. From these data, it could be estimated that since the industrial revolution, the Cd content of soils may have doubled due to anthropogenic influence, suggesting a 50/50 ratio for the natural/anthropogenic Cd content of agricultural soils.
On the other hand, the anthropogenic emissions to air have been significantly higher in former times, e.g. the 1950s, 1960s, then at present. Consequently, there is no direct relationship between the atmospheric emission pattern of the 1990s (Table 1), and the anthropogenic inputs through atmospheric deposition since the previous century. The ratio between natural and anthropogenic soil Cd, used in this analysis (20/80), should therefore be considered with great caution and most probably represents a worst case generalisation. It is clear that this ratio is strongly influencing the end result of the analysis, taken into account the relative importance of the soil-plant-food transfer for human exposure.

Step 4 :
Anthropogenic sources of Cd to soils are: P-fertiliser application, atmospheric deposition and application of sewage sludge in a 60/30/10 ratio (based on Jenson & 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, ad after deposition on : Cd in soil (30% of total input to soils), Cd in exposed plant parts (30% of total plant Cd content), 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 water effluents and surface waters results in sludges that can be applied to agricultural soils. In 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 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 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 50 ng/L (EU RA, FOREGS, 2004). The EU Cd RA refers to an average 90P across monitoring rom 7 countries at 160 ng/L. The contribution of natural Cd in surface waters is thus estimated at 30%.
- 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 cadmium industry was revised downward to 215 kg, taking into account recently reported emissions from 2003 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 0.98 = 16.13% +
deposition on plants : 0.3 x 0.98 x 0.98 = 28.21% +
direct inhalation = 2% +
sludge to soil : 0.15 x 0.5 x 0.7 x 0.1 x 0.8 x 0.7 x 0.98 x 0.98 = 0.28% +
deposition to water (cf Table 2) = 0.55%
= 47.8%

b) Sources to water
Sludge to soil : 0.5 x 0.7 x 0.1 x 0.8 x 0.7 x 0.98 x 0.98 = 1.88% +
Direct to aquatic food : 0.65 x 0.01 x 0.98 = 0.637% +
Direct to drinking water : 0.65 x 0.01 x 0.98 = 0.637%
=3.2%

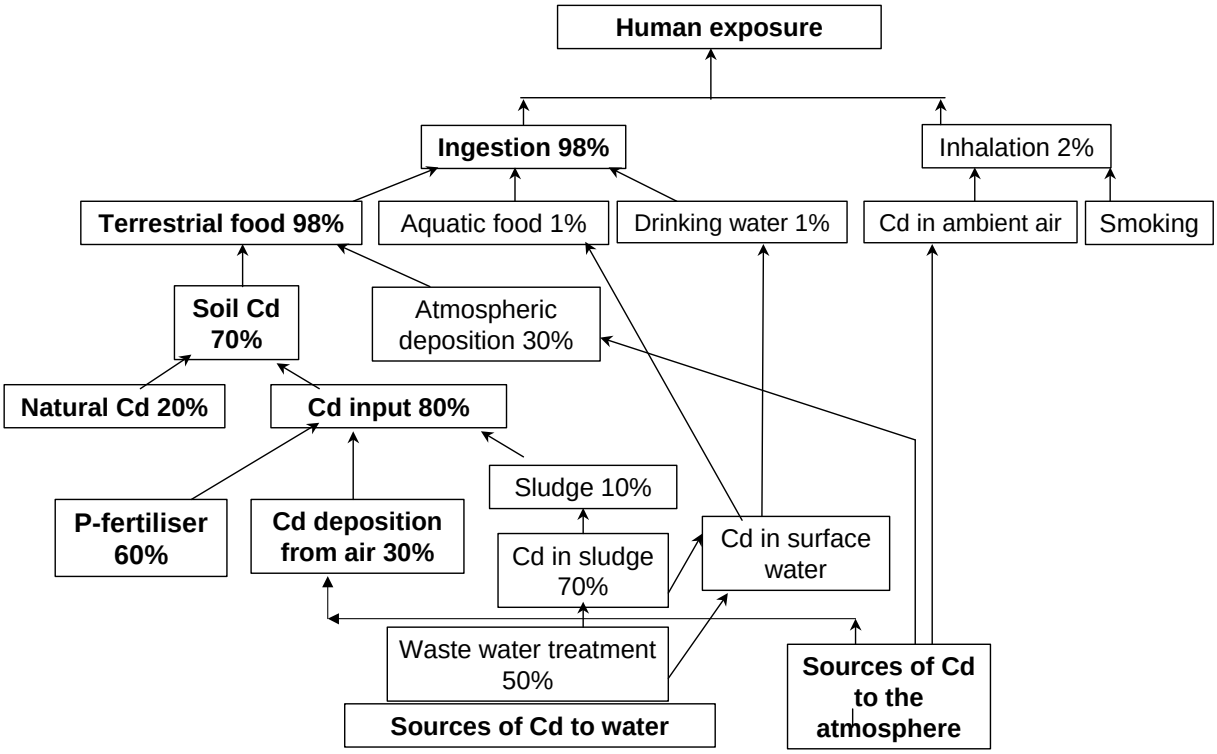
c) P-Fertiliser direct to soil
0.6 x 0.8 x 0.7 x 0.98 x 0.98 = 32.27 + P-fertiliser to water term

d) Natural Cd
To soil : 0.2 x 0.7 x 0.98 x 0.98 = 13.4% + 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 a 47.8% contribution from atmospheric sources to total exposure, and a contribution of 3.2% from water sources to total exposure: Table 2.

Combination of the information from a) to e) results in the following end result (see also Figure 2) :
- natural sources : 19.7% (13,4% to soil + 5.3% to air + 1% to water)
- P-fertiliser : 32.7 (application to soil : 32.3% + production to water : 0.4%)
- fossil fuel combustion : 19.6% (to air)
- iron and steel production : 11.7 (to air : 11% + water : 0.7%)
- non-ferrous metals production : 4.2% (3.8% to air + 0.4% to water)
- production and use of Cd products : 0.3% (to air)
- other (cement manufacturing, traffic,…) : 6.7% (to air)
    - municipal waste incineration : 1% (to air)

Figure 1. Pathways of human exposure to Cadmium



The relative contribution of different sources to Cd exposure of general populations

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Contents

- Cd emissions to air and water
- General population exposure to Cd:
  - Quantitative analysis of Cd transfer pathways
  - Main sources of Cd transfer to food
- Specific case
  - Smokers
- Conclusions

Sources of Cd to air and water (EU; T/y)

	air	%	water	%
Natural sources	15	11	19	20
Combustion of fossil fuels	55.7	41	n.s.	
Iron and steel production	31	23	21.2	22
Non-ferrous metal production	11	8	11.0	12
Production/use Cd products	1.2	1	1.1	1
Production P-fertilisers	0.7	0.5	25.7	27
Waste incineration	3.2	2	1.6	2.2
Other (cement manufacture, traffic,…)	19	14	1.7	2.4
Atmospheric deposition	-	-	16.7	18

How do Cd-sources to air and water translate into Cd-exposure of the general population?

Critical pathways of human exposure to cadmium (non-smokers)

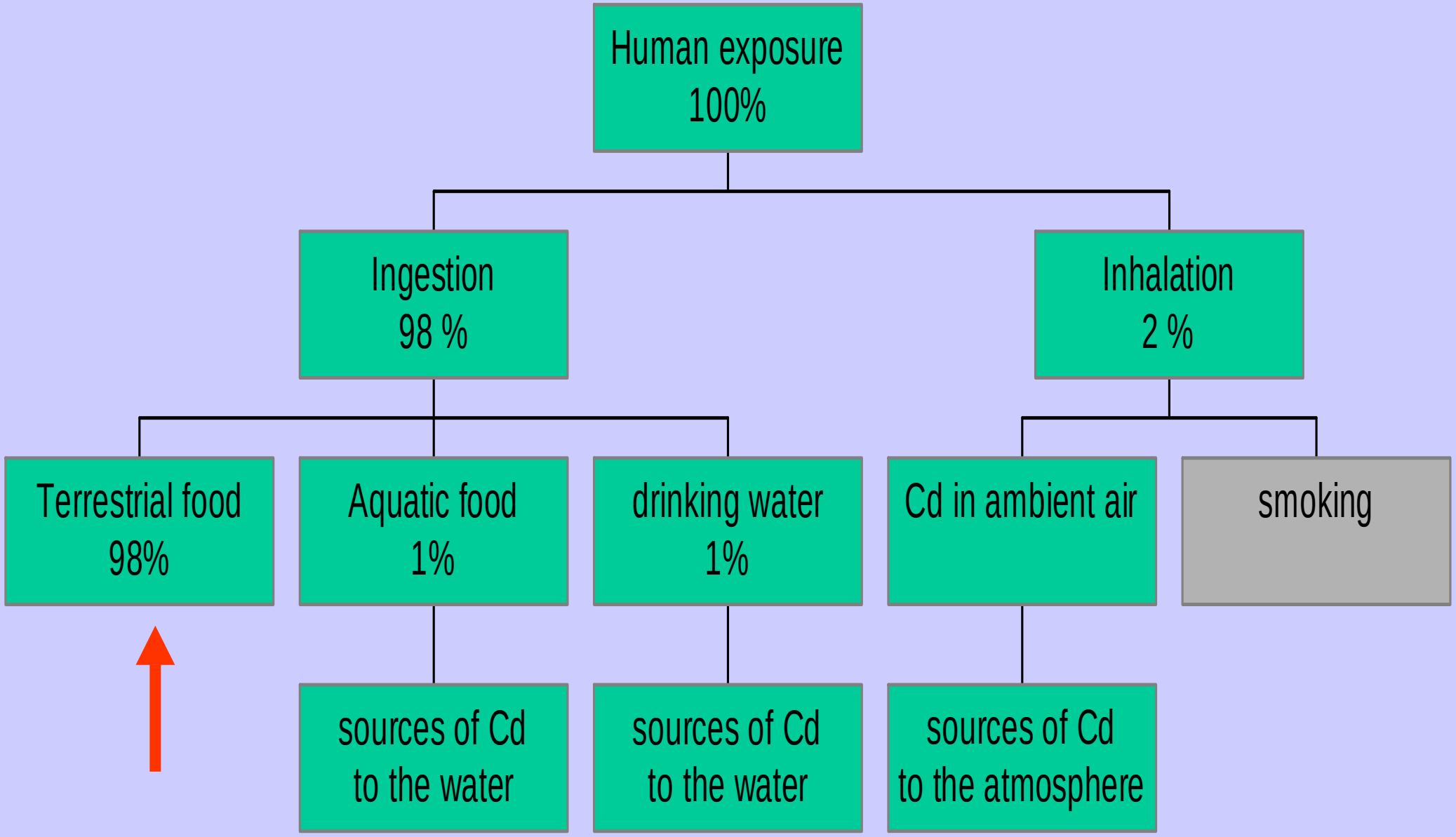
- Inhalation:
  - 5 ng/m^3 x 12 m^3/d = 60 ng/d x 50 % = 0,03 µg/d
- Ingestion:
  - 30 µg/d x 5% = 1.5 µg/d
- Total internal dose: 1.53 µg/d

  Inhalation: 2%

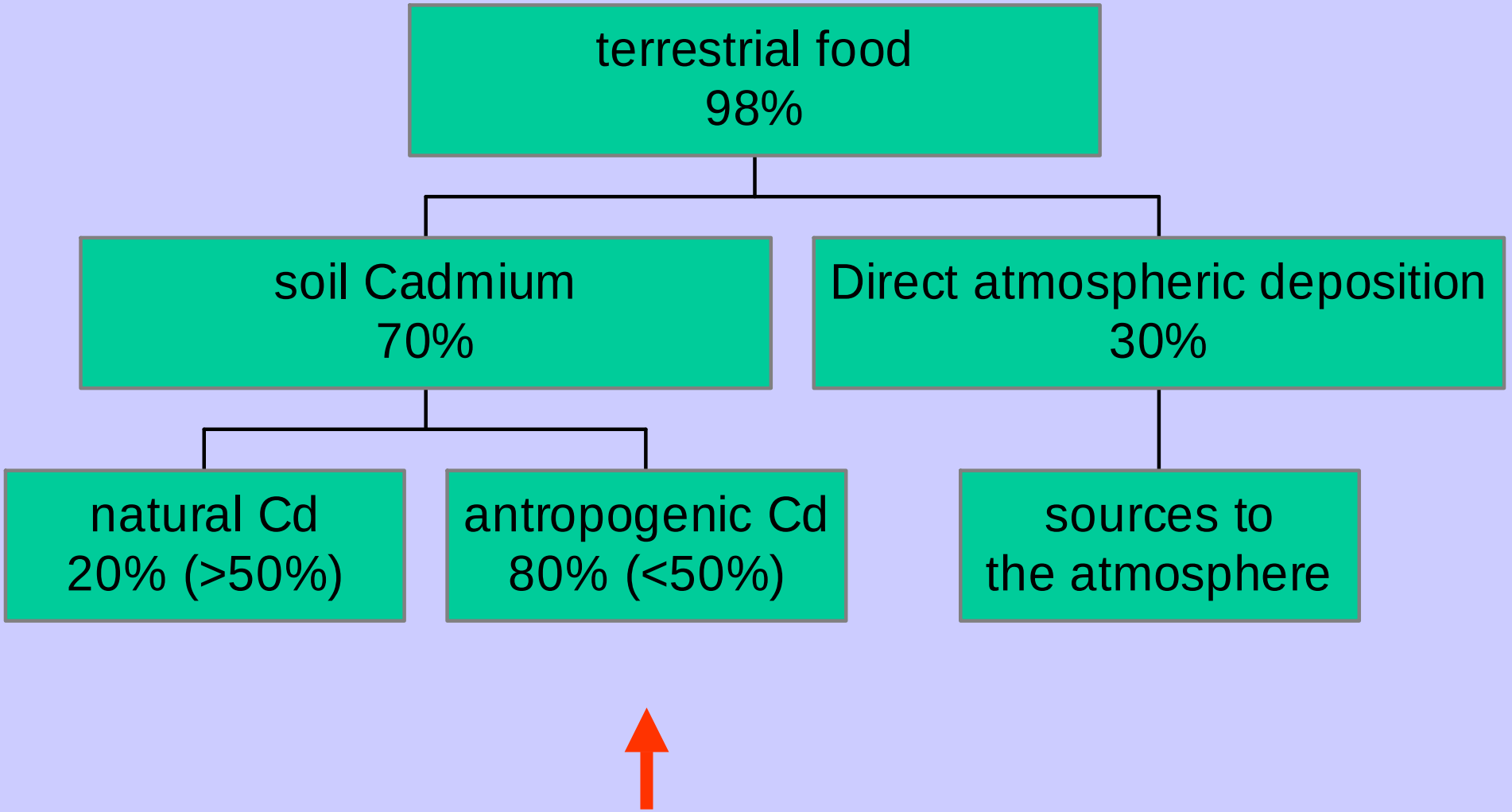
  Ingestion: 98%

  Ingestion:
  - > 98 % « terrestrial » food
  - < 1 % « aquatic » food

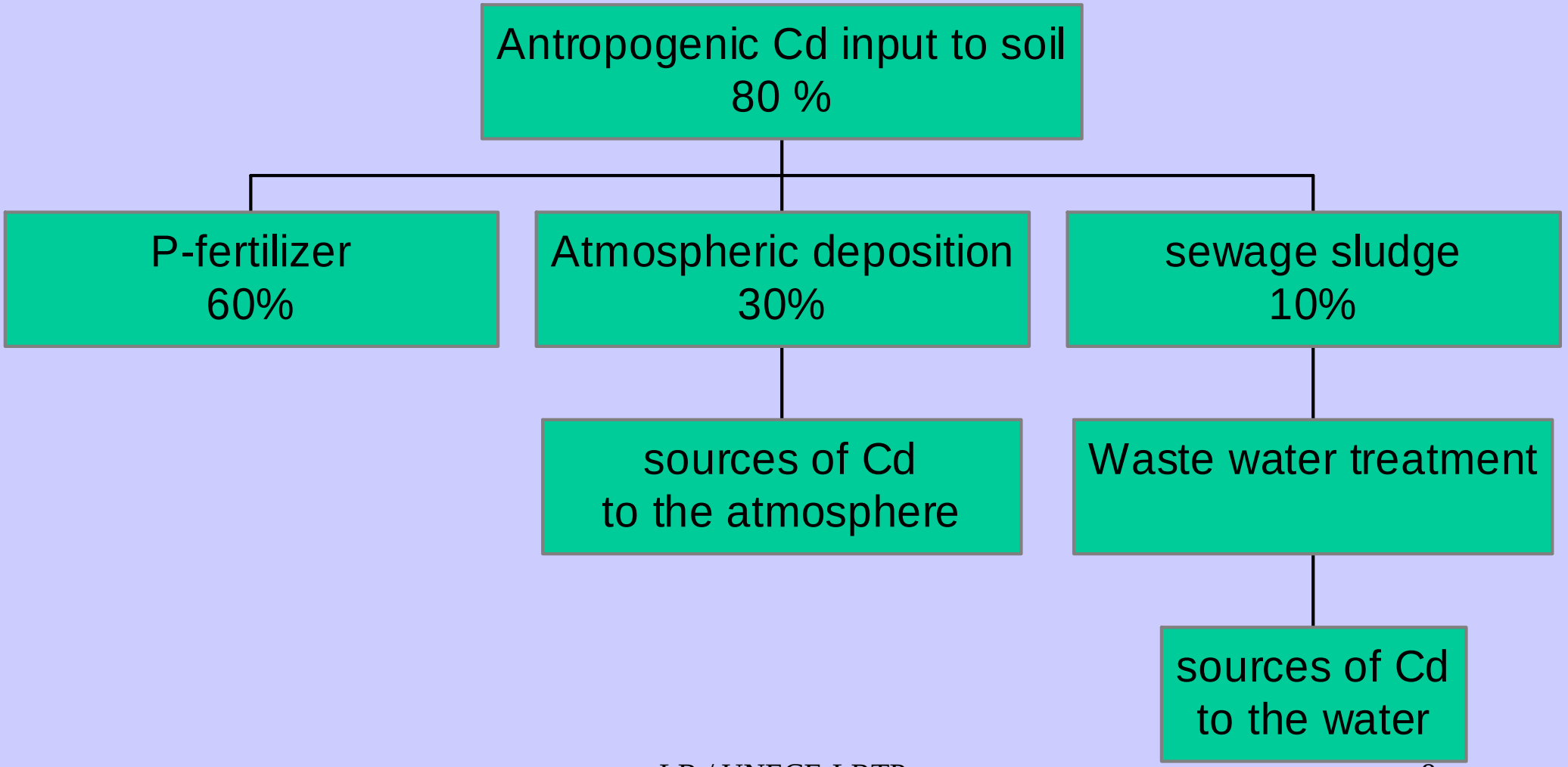
Critical pathways of human exposure to cadmium (1)



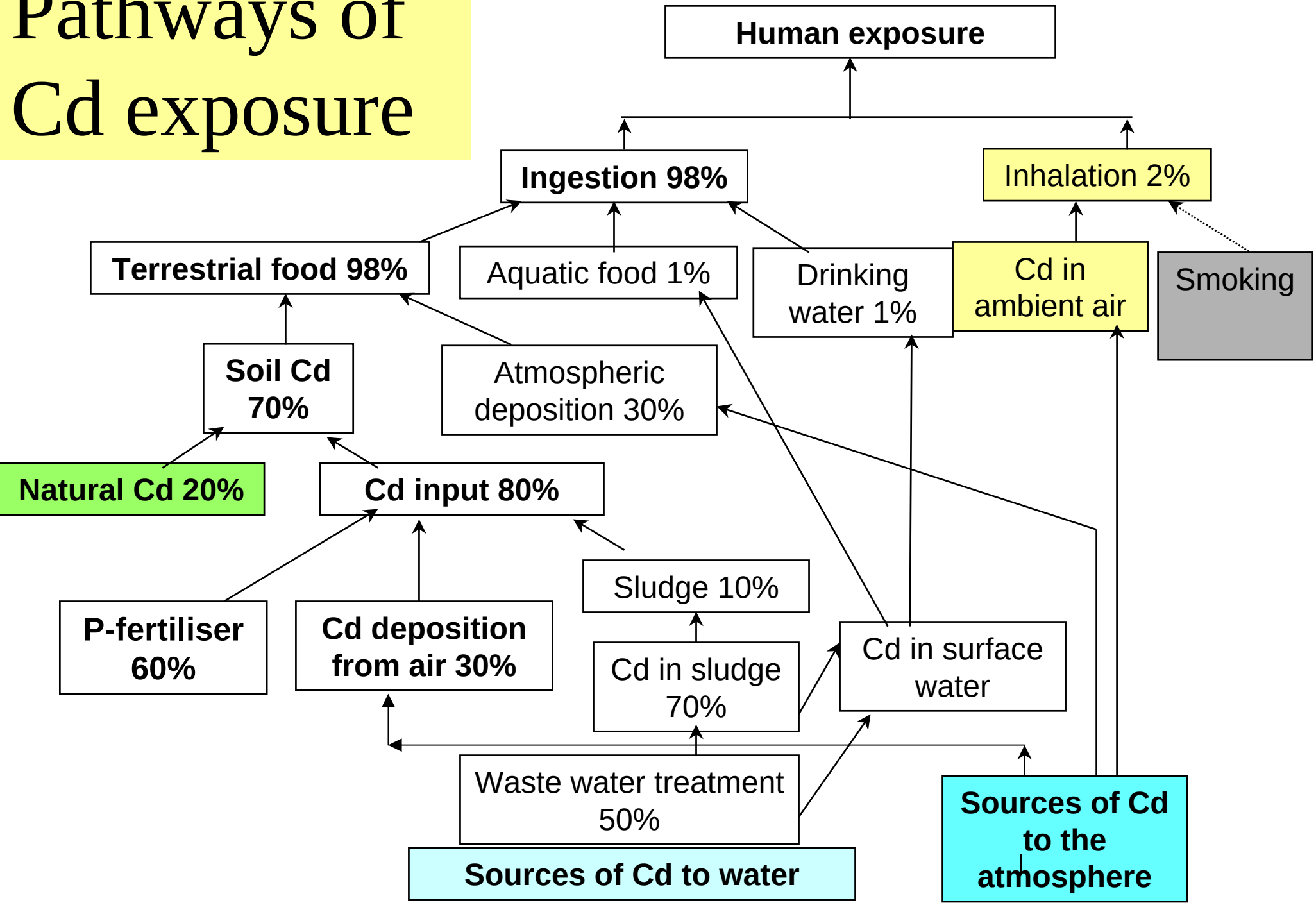
Critical pathways of human exposure to cadmium (2)



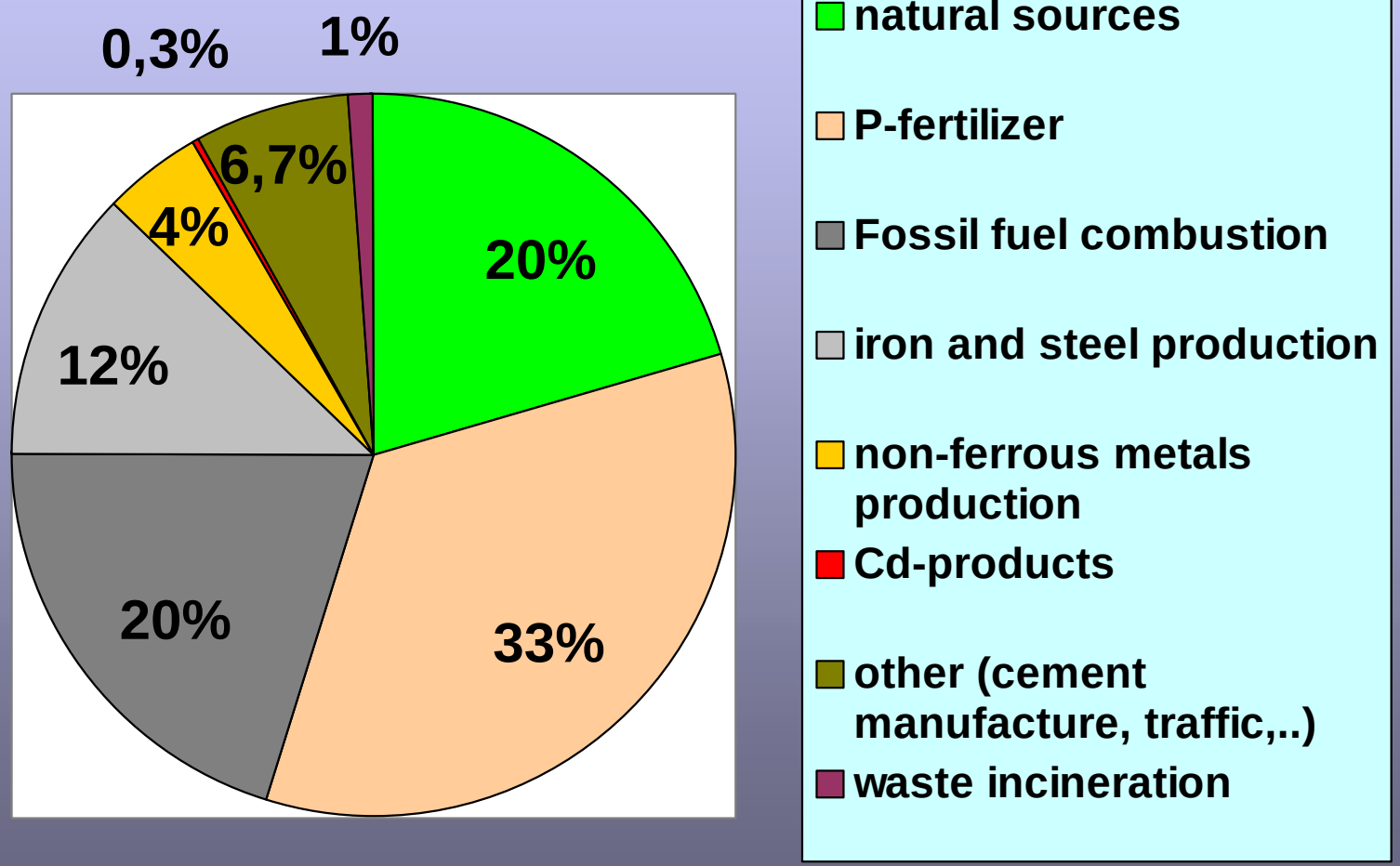
Critical pathways of human exposure to cadmium (3)



Pathways of Cd exposure



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



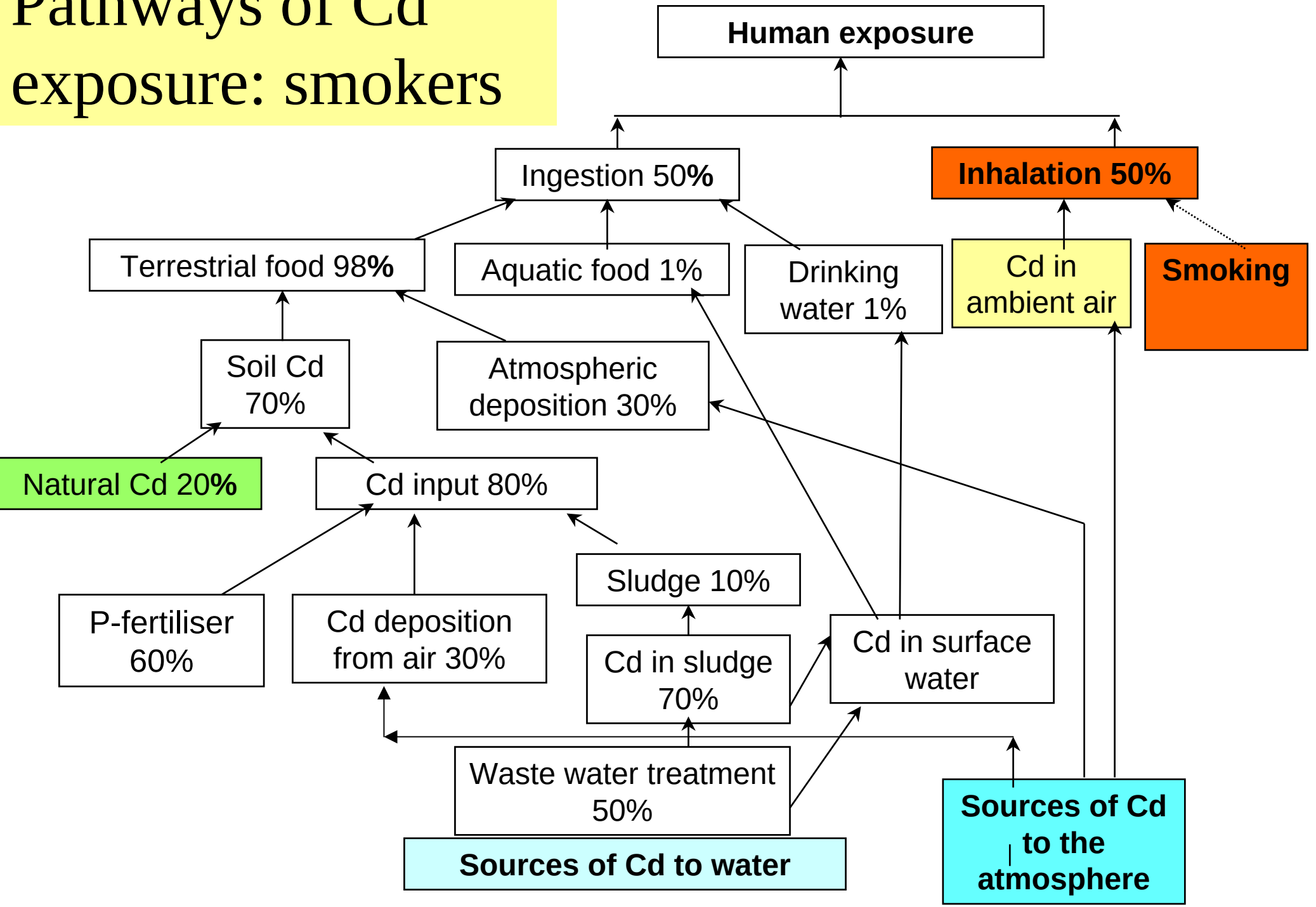
Factors affecting the relative contribution pattern

- *smoking habits*
- input/content of P-fertiliser to soil
- vicinity of strong atmospheric emission source
- living in (historically) contaminated area: drinking water, soil Cd
  - Local soil conditions
- specific dietary habits

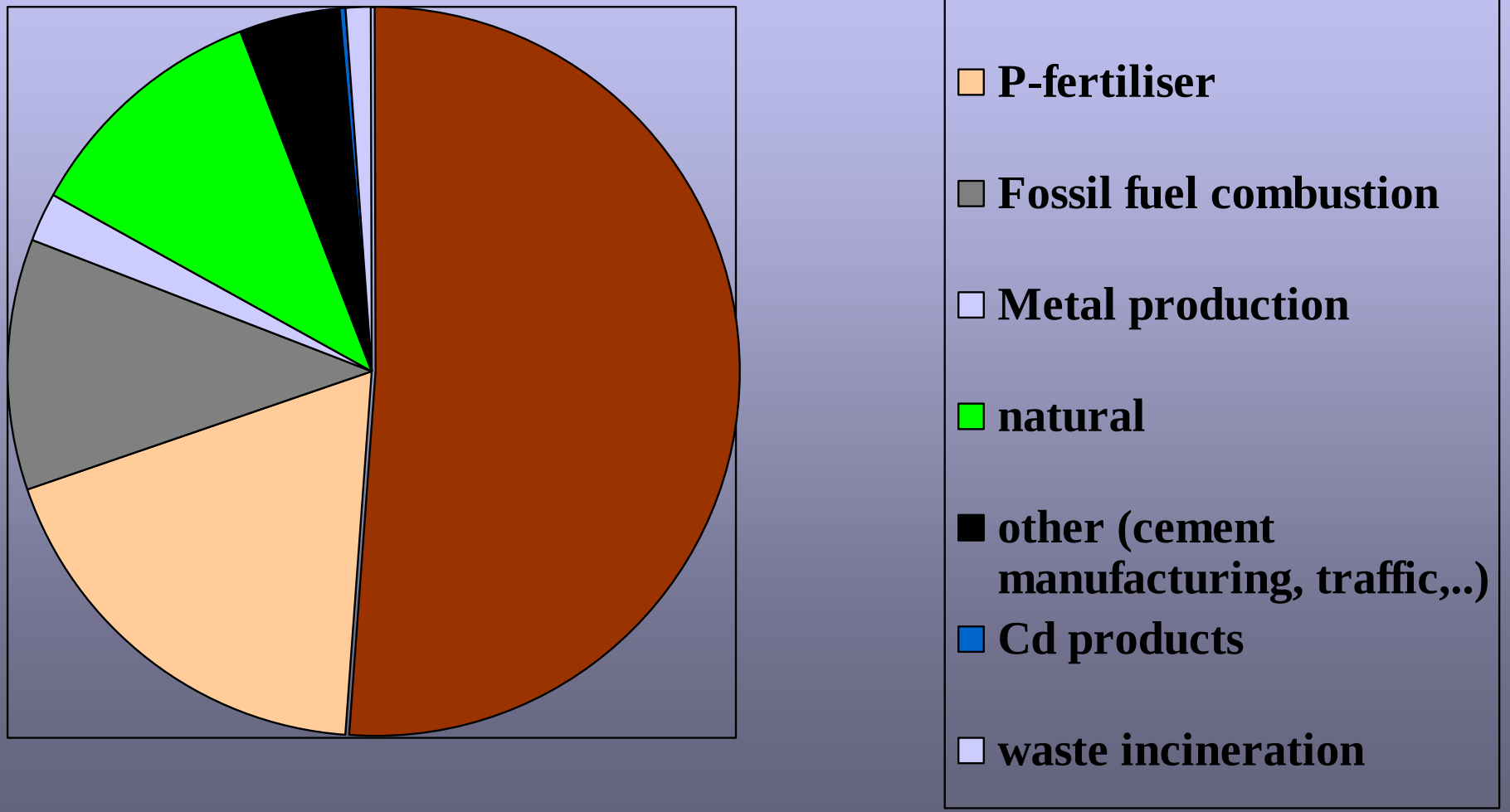
Specific case: smoking

- Smoking 20 cigarettes/d x 1,5 µg/c = 30 µg/d
  - 10 % inhaled = 3 µg/d
  - 50 % absorbed = 1,5 µg/d
  - Total dose: 1,5 + 1,5 µg/d = 3 µg/d
- Exposure due to smoking: 50% of total
- Passive smoking:
  - Cd Air indoor: 100 ng/m3
  - 12 hrs exposure = 1,2µg/d x 0,5 = 0,6µg/d (~ diet)

Pathways of Cd exposure: smokers



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



Conclusions

- The transfer pathways of Cd from the environment to man are well documented
  - this allows to calculate the relative contribution from different Cd-sources to human exposure
- In the general environment
  - Direct inputs to soil and air emissions are more important then emissions to water
  - Natural Cd is significant
  - The contribution from Cd-products is negligible
- Smoking and dietary habits influence significantly the transfer pattern, and well as local contaminations
  - Population exposure monitoring should be basis for assessment