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Lead in the Dutch environment:
a review of exposure pathways and dispersion models
A pilot study for an integrated chain model

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SUMMARY

Prognoses of possible effects and risks of chemical emissions into the environment and calculation of the effects of countermeasures are often produced using mathematical models. Most models are developed to describe a part of the total source-to-effect chain or to give results for specific compartments. In this study the models available at the RIVM have been examined for their usefulness in an integrated chain model for the total source-effect-risk chain of lead and lead-210 (a radioactive isotope of lead, $^{210}\text{Pb}$) in the Dutch environment. An integrated chain model is defined here as a set of relatively complex, deterministic stand-alone models, which have been independently developed and later linked.

Lead can have serious effects on both human health and the environment. This report presents a short survey of the emission sources and effects of lead. Inorganic lead is emitted to the atmosphere in rather large quantities by traffic and industrial plants. $^{210}\text{Pb}$ is emitted to air mainly by brickworks and phosphate plants. For human exposure inorganic lead and $^{210}\text{Pb}$ emitted to the atmosphere form an important starting-point for most of the pathways through which humans are exposed to lead. The most important pathways are inhalation, external irradiation (for $^{210}\text{Pb}$ only), deposition on open water or agricultural lands, followed by consumption of resulting produce such as mussels, milk and meat.

Although the direct emission of lead to water and soil can not be neglected, the total load of (inorganic) lead on the human body originates mainly from atmospheric emissions. On the basis of an overview of the processes involved in modelling the source-to-effect chain, models are selected for their ability to describe the above-mentioned pathways.

The overview on pathways and models indicates that the sources of lead are both point and diffuse, whereas the sources of $^{210}\text{Pb}$ are merely point. In radiation protection, doses are calculated for the most exposed individuals, implying that ecosystems are also fully protected, whereas for lead, exposure limits are used for protection of both humans and ecosystems. Due to these differences, models for lead are to be used on local, regional and national scales, while for $^{210}\text{Pb}$ the use of models is restricted to local scale. However, the approach to establishing an integrated chain model will be the same for lead and $^{210}\text{Pb}$.

The source-to-effect chain is only partly covered by the available models. On a local scale the incompleteness is restricted to relatively small parts of the chain, like the contamination and transfer of lead from groundwater to terrestrial plants. On a regional scale also models for the distribution pathways from water to soil and from water to aquatic organisms are missing. On a national scale only models describing dispersion pathways from atmosphere to water and soil and those for the calculation of the dose from
radioactive lead in humans are available.

The development of an integrated chain model is very time consuming, therefore should only be developed if there is a specific demand. Hence it would be advisable to start a follow-up project with an inventory of the demands for integrated models to answer specific questions on lead and to determine what is required: either the development of an integrated chain model or a less detailed but more encompassing integral model.
SAMENVATTING

Bij het opstellen van prognoses van de mogelijke effecten en risico's van emissies van stoffen in het milieu, en de berekening van de effecten van tegenmaatregelen wordt vaak gebruik gemaakt van modellen. De meeste modellen zijn ontwikkeld om bepaalde compartimenten of delen van de totale bron-effect keten te beschrijven.

In deze studie zijn de modellen die aanwezig zijn op het RIVM onderzocht op hun geschiktheid een geïntegreerd ketenmodel voor lood en radioactief lood (\(^{210}\text{Pb}\)) in het Nederlandse milieu te vormen. Een geïntegreerd ketenmodel bestaat uit een set complexe, deterministische, zelfstandige modellen. Deze zijn onafhankelijk van elkaar ontwikkeld en later met elkaar verbonden.


Voor de blootstelling van de mens vormen emissies van anorganisch lood en \(^{210}\text{Pb}\) naar de lucht veelal het beginpunt.

De bronnen van lood zijn zowel lokaal als diffuus van aard terwijl die van \(^{210}\text{Pb}\) voornamelijk lokaal zijn. De belangrijkste blootstellingswegen zijn daarbij: inhalatie, externe bestraling (voor \(^{210}\text{Pb}\)) depositie op oppervlaktewater of landbouwgrond, gevolgd door de consumptie van de hiervan afkomstige produkten zoals mosselen, melk en vlees. Hoewel de directe lozing op bodem en oppervlaktewater niet mag worden verwaarloosd, is de totale lichaamsdosis direct of indirect voornamelijk afkomstig van via de lucht in het milieu gebracht lood.

In de stralenbescherming wordt de maximale dosis opgesteld voor de meest blootgestelde personen, waarbij impliciet wordt aangenomen dat daarmee ecosystemen voldoende beschermd zijn. Voor niet-radioactief lood wordt gebruik gemaakt van limietwaarden die expliciet bescherming bieden aan mens en ecosystemen. Deze verschillen houden in dat er voor lood in principe zowel op lokale, regionale als nationale schaal modellen opgesteld worden, terwijl voor \(^{210}\text{Pb}\) volstaan kan worden met modellering op lokale schaal. Desondanks is de benadering van het opstellen van een geïntegreerd ketenmodel voor lood en \(^{210}\text{Pb}\) gelijk.

Voor het geïntegreerde ketenmodel zijn modellen geselecteerd die de bovengenoemde processen beschrijven.

De bron-effect keten kan slechts gedeeltelijk worden beschreven met de aanwezige modellen. Op lokale schaal beperken de hiaten zich tot betrekkelijk kleine delen zoals de overdracht van grondwater naar terrestrische organismen. Op regionale schaal zijn ook modellen voor de verspreiding van water naar bodem en van water naar aquatische organismen afwezig. Op nationale schaal zijn slechts modellen aanwezig die de verspreiding van lucht naar water en bodem, van water naar de mens berekenen, en
modellen die de stralingsdosis berekenen ten gevolge van blootstelling aan $^{210}$Pb.

De ontwikkeling van een geïntegreerd ketenmodel is een tijdrovend en dus kostbaar proces. Het is derhalve wenselijk pas over te gaan tot het ontwikkelen van een dergelijk ketenmodel, wanneer is gebleken dat te beantwoorden vragen het best met een dergelijk model beantwoord kunnen worden. Daarom wordt geadviseerd een vervolgproject te starten met de vaststelling van de vragen die met behulp van het ketenmodel beantwoord moeten worden. Dan valt vast te stellen of hiervoor het best een geïntegreerd ketenmodel dat gedetailleerde informatie oplevert gemaakt kan worden, of een integraal model dat minder gedetailleerd is maar eenvoudiger en sneller.
1 INTRODUCTION

One of the tasks of the National Institute of Public Health and Environmental Protection (RIVM) is to describe the state of the environment (diagnosis and prognosis) and the distribution of chemical agents and compounds through this environment.

To predict the possible effects and risks of emissions of chemicals, models describing the source-to-effect chain are developed. Most of these models are developed to describe parts of the chain and give results that are oriented towards specific compartments or parts of the chemical flow. These models are constantly adapted and improved as new research questions arise. Consequently, these models can not be easily linked, and in- and output data are often not compatible.

The aim of this study is to identify the gaps between the available models in order to come to a chain model for lead.

The report describes to what level (and how) the knowledge of the total source-to-effect chain from emission → distribution → exposure → effect → risk for lead (symbol Pb) is implemented in the models that are available at the RIVM. It provides an overview of the available models that can be used for lead and the compatibility of the in- and output data of the selected models.

The selection of lead as a pilot for the construction of a chain model has been based on the following arguments:
1. Lead is an important chemical agent from a substance-related environmental policy point of view;
2. Several laboratories at the RIVM use different models for describing (parts of) the chain concerning lead;
3. From an environmental point of view lead is not only of importance as a heavy metal but also as a radionuclide ($^{210}$Pb).

In chapter 2 a general introduction on models describing the source-to-effect chain is given. To give an impression of what sources, processes, pathways and effects play a role in the chain from emission to effect, a compilation of the available knowledge concerning the actual lead emission, possible exposure pathways and effects is presented in chapter 3. The most important sources, pathways and effects are abstracted that must be included in the models to cover the chain model. In chapter 4 the available models are described. The gaps in the total set of models and discrepancies between the different in- and output formats are identified and described in chapter 5. Finally, in chapter 6 the conclusions and recommendations from this study are formulated.
The project is initiated by D. van Lith, and conducted under the coordination of the Laboratory of Radiation Research (LSO). The members of the project group were: GJ Eggink (LSO) and PAM Uijt de Haag (LSO), JA Annema (Laboratory of Waste and Emissions, LAE), LC Braat (Environmental Forecasting Bureau, MTV) and JJM van Grinsven (Laboratory of Soil and Groundwater Research, LBG).

Besides the above mentioned persons, experts from the Toxicology Advisory Centre (ACT), Air Research Laboratory (LLO), Laboratory of Ecotoxicology (ECO), Laboratory of Water and Drinking-water Research (LWD), and the Centre for Mathematical Methods (CWM) have been consulted.
2 MODELLING THE SOURCE-TO-EFFECT CHAIN: TWO DIFFERENT APPROACHES

2.1 Introduction

To calculate the distribution, the effects and the risks on human health or ecosystems of chemicals and to calculate the effects of measures models describing (parts of) the source-to-effect chain are developed. Some of these models describe specific compartments in detail, while others cover the whole source-to-effect chain in a less detailed way.

For a description of the whole source-to-effect chain one can either use models covering the whole source-to-effect chain, or create a chain model by linking some of the detailed models, and adjust the in- and output data. Both types of source-to-effect chain models have their own advantages and disadvantages.

2.2 Integrated modelling

The first and second National Environmental Outlook reports [RIVM88, RIVM91] were the result of scientific exercises with a great number of quite heterogeneous environmental models. Most of these models were developed and residing in the RIVM, some were external. Their signature can be characterized as ranging from simple regression models to complex process descriptive and explanatory models. At that time, these models were generally considered to be "state of the art and science", i.e. the best available representations of the structure and processes in major environmental compartments and receptor systems. This ensemble of models, and their recent updates, are quite effectively used in analysis and prediction of dynamics and future states in the parts of the environment they represent.

Models describing the entire chain from source to effect (and risk) can be built by adjusting the in- and output of the submodels. Such a new model usually consists of linked complicated models, now treated as modules in a new chain model. These modules describe each a small part of the source-to-risk chain, such as the dispersion of substances in the atmosphere, the chemical speciation in the soil or dose effect relationships. Since these models are complex, they require relatively large computing times and their application is usually limited to a single scenario and a single source.

There are various interpretations of the concept integrated chain model. Often it is understood to be a model which includes and links modules (or just a few equations) from two or more scientific disciplines. The major disciplines involved in environmental policy analysis are: economics, process technology, air chemistry, geography, hydrology, soil science, ecology, agro-ecology, human health sciences, while many applications further require input from computer and information science, human psychology and decision
In this document an integrated chain model is defined as a set of stand-alone models describing the whole source-to-effect chain, with interface routines, and data transfer protocols for data transfer between RIVM laboratories.

The efforts in creating an integrated chain model concentrated first on the development of *interface routines* between the existing detailed environmental models. This approach was characteristic for the MilMan concept. The MilMan (MILieu (= environmental) MANagement) concept would involve a RIVM wide effort with the objective to produce an Integrated Environmental Management System. The MilMan system was designed to include detailed process models, interface routines, and protocols for data transfer. The idea behind this approach was that a shorter run-time for a scenario-analysis could be achieved if the output data of a certain (source) model to another (receptor) model in the chain are formatted according to requirements of the receptor model. Several initiatives were taken to explore the feasibility (e.g. the project of which the present report is the result), discussions were held but the concept never became a formal project.

### 2.3 Integral modelling

In another approach, the focus was shifted from a set of stand-alone models towards a single comprehensive environmental model, which should be assembled through integration of relatively simple modules. These models should be based on detailed process models and represent environmental compartments and receptor systems. This type of model is called an *integral chain model*. In this type of model the results of experiments and the results of calculations with complicated models are often used as simple relationships. Since these models require relatively little computing time they are used to calculate a vast number of scenarios for one or more policy themes.

At RIVM a model of this type with the associated user interface, control program, database, database management system and output generator is EXPECT (EXPloring Environmental Consequences for Tomorrow). The modules in EXPECT are developed as simplified versions of existing detailed compartment models, or, if these are not available yet, as temporary simple precursors.

The philosophy behind the EXPECT approach is that quality control is better and speed of calculations is greater with a model which is housed in a homogeneous software and hardware environment. Data flow control is part of the EXPECT system so that separate interface routines are not required in this approach.
2.4 The organization of integrated modelling

Figure 1 illustrates the organizational position of the two approaches and associated models and databases within the National Institute of Public Health and Environmental Protection.

<table>
<thead>
<tr>
<th>DATA</th>
<th>MODELS</th>
</tr>
</thead>
<tbody>
<tr>
<td>aggregated</td>
<td></td>
</tr>
<tr>
<td>Environmental core</td>
<td>EXPECT</td>
</tr>
<tr>
<td>database MILKERN</td>
<td></td>
</tr>
<tr>
<td>detailed</td>
<td></td>
</tr>
<tr>
<td>RIVM laboratory</td>
<td>RIVM (SVII)/MiiMan</td>
</tr>
<tr>
<td>databases</td>
<td>models</td>
</tr>
</tbody>
</table>

**Figure 1** Organizational position of models at the RIVM

The lower left box represents the detailed environmental data available within the RIVM. They are obtained through monitoring, field studies and laboratory research, as well as acquired from other research organizations. These data are used by the detailed process models (shown in the lower right box) most of which are housed in the laboratories of Sector VII of RIVM. The upper left box indicates the environmental core database (MILKERN) within the Environmental Forecasting Bureau. The EXPECT model system (see the upper right box) is developed in line with the MILKERN database.

The EXPECT system is dependent on the base models which are the components in the MilMan concept. The main difference is that in the MilMan concept the integration is engineered after independent model development, while the EXPECT system is developed from an integrated design before the constituting modules are developed. The models which are the basis of EXPECT modules are expected to change and be updated. Through the RIVM network of modelling teams and the yearly update of the catalog of environmental models at RIVM, such information is relayed.

The current state of affairs is that after two years of design and implementation, the EXPECT system is operational for a limited number of assessment types, while a great number of modules are under construction both at the laboratories of Sector IV and Sector VII. Detailed process models are still being used for Environmental Outlooks as well as for their original purpose, compartment policy or environmental policy theme assessments. Also, some of the EXPECT modules will be used for such specialized assessments.
3 LEAD IN THE DUTCH ENVIRONMENT: FROM SOURCE TO EFFECT

3.1 Introduction

Lead and its compounds are widely distributed in nature, and lead is also continuously discharged into the environment due to human activities and the radioactive decay of radium-isotopes. In order to assess the consequences of the presence of lead in the environment and the discharges of lead into the environment, model calculations can be performed. These model calculations cover the entire chain from the discharge of the source and the dispersion in the environment to the exposure, effects and risks to both humans and ecosystems. However, large differences exist between the various sources of lead and the resulting effects depend upon the chemical species of lead emitted by the source. Consequently several models are required, the choice being dependent on the characteristics of the source and the chemical species of the discharged lead.

A short survey of the distinct sources and effects of lead and its compounds is given in this chapter. In chapter 3.2 the most important sources of lead are reviewed along with the relevant characteristics; the main exposure pathways and effects to humans and ecosystems are presented in chapter 3.3. This information is necessary for the formulation of the demands on an integrated source-to-effect chain model for lead. For a more comprehensive survey of the distribution of lead in the environment and the effects of lead the reader is referred to the reports of Janus et al. and the CCRX [Ja91, Kr90].

Not only the effects in humans, but also the effects in animals and ecosystems are studied. In the policy of the protection of an ecosystem the starting point is the protection of species and the protection of functions of an ecosystem [VROM89]. It is assumed that the protection of the species (both qualitative and quantitative) in an ecosystem warrants the protection of the functions of an ecosystem. In environmental policy, species are protected at population level. Protection of the ecosystem as a whole is therefore restricted to protection of a population.

Based on differences in effects on to humans, it is standard practice to distinguish three different chemical species of lead: inorganic lead, organic lead and metallic lead. Besides the various chemical species, the radioactive isotopes of lead have also been identified as harmful to humans due to the carcinogenic properties. Since the radioactive isotope $^{210}$Pb is abundant in nature, the consequences of the emission and distribution of $^{210}$Pb have to be assessed.
3.2 Quantitative treatment of lead sources

Due to human activity, approximately 12,000 tons of lead are introduced into the Dutch environment annually (figures for the year 1989, [Ja91]). The total amount of lead emitted to the Dutch environment has decreased in the last decade, mainly due to the use of unleaded fuel, and a further reduction is foreseen [Ja91]. An overview of the different sources of lead and their emission to the different compartments is given in figure 2.

![Diagram of lead sources and compartments](image)

Figure 2 Total discharges of lead to the environment in the Netherlands. Figures are given for the year 1989 in tons per year [Ja91]. Both the discharges to the total soil are given as well as the discharges to agricultural land (in parentheses). Deposition of lead from sources abroad is included in the figures, and amounts to 77% of the total deposition (in 1989).

3.2.1 Industrial plants, power stations, incinerators and sanitary landfills

The lead emissions to the atmosphere by power stations, incinerators and industrial plants, consist mostly of inorganic lead compounds (lead oxides and lead-sulphur compounds), which are bound to aerosols. The most important industrial source of lead emission to the atmosphere is the steel producing industry (approximately 48 tons lead in 1989 [Ja91]). Lead is also emitted to the atmosphere by coal-fired power stations by coal combustion (approx. 1 ton per year, 1989 [Ja91]). The remaining combustion products are to a large extent reused and only a small part (containing approx. 0.4 ton lead) is stored.

Industrial emissions to water are dominated by the fertilizer industry by the processing of
phosphorous ore (approximately 12 tons lead in 1989 [Ja91]).

Industrial lead-containing waste (approx. 6660 tons, 1989) is either stored or used in landfills. Emission of lead into the soil and groundwater occurs in case of leaking or absent liners. Measured values of lead in landfill leachate in the Netherlands range from 0.0 to $3.0 \cdot 10^4$ µg per litre leachate. However, measured values are only available for lined landfills with leachate collection systems, and the values from older, closed landfills are unknown. Using averaged values for lead measured in leachate, 5-hectare landfill dimensions, average infiltration and a total of 4000 landfills in the Netherlands, a very generalized value of 4 tons lead per year can be assumed. This value is applicable only to lined landfills, however, which are rigidly controlled [Po92].

3.2.2 Road traffic

The main contribution to the air concentration of lead in 1989 was the use of leaded petrol in road traffic. Lead is added to petrol as organic lead compounds. However, in the combustion process the organic lead is converted into inorganic lead [DHSS80]. Hence the emission to the air of the road traffic consists to a large extent of inorganic lead, mostly bound to fine particles. The particle size is very small: 80-98% is bound to aerosols less than 1 µm and only 0.1-10% is bound to particles larger than 10 µm [Kr90]. Only a small part of the lead in petrol is emitted as organic lead, due to evaporation of petrol and unburned compounds in the exhaust; in the atmosphere, organic lead is also (finally) decomposed into inorganic lead. Hence the amount of organic lead in the total air concentration is very small under normal conditions. However, for example near petrol stations, the air concentration of organic lead is not negligible.

3.2.3 Fertilizer, manure, sewage sludge, compost

The use of fertilizer, manure and sewage sludge in agricultural practice results in a diffuse contamination of agricultural land with inorganic lead compounds.

Compost is made either out of sludge from sewage treatment plants or out of municipal waste (total 25 tons of lead in 1989, [Ja91]). Compost is used entirely by individuals and in public green areas and therefore results in a contamination of the soil in build-up areas. The use of compost in allotment gardens results in the contamination of crops for private consumption.

3.2.4 Hunting, fishing, skeet-shooting

The use of lead in hunting, fishing and skeet-shooting results in large emissions of lead to the environment and the contamination of soil and surface water with metallic lead. The contamination is either restricted to the areas around the rifle-range in the case of target
shooting, or dispersed over a larger area in the case of hunting. The use of lead in sinkers results in a contamination of surface waters and sediments with metallic lead. Approximately 20% of the emission occurs to inland waterways whereas the remaining 80% is emitted to sea waters. Due to weathering and chemical interaction metallic lead is converted to inorganic lead in the soil and water.

3.2.5 Import

Lead emission in neighbouring countries is an important source of contamination for Dutch environment. No data are currently available on the contribution of sources abroad to the air concentration of lead in the Netherlands. It is estimated that approximately 77% (1989) of the total deposition of lead originates from sources abroad. Another important source of lead is the transport of contaminated sediment in the large rivers (Rhine, 243 tons lead, Meuse 35 tons, Scheldt 40 tons, 1989). The use of contaminated river sludge for raising land levels results in a diffuse contamination of the soil.

3.2.6 Radioactive lead ($^{210}$Pb)

The decay chains of the primordial radionuclides uranium-238 ($^{238}$U), uranium-235 ($^{235}$U) and thorium-232 ($^{232}$Th) comprise several radioactive isotopes of lead: lead-214 ($^{214}$Pb, half-life 27 minutes), lead-212 ($^{212}$Pb, 11 hours), lead-211 ($^{211}$Pb, 36 minutes) and lead-210 ($^{210}$Pb, 22 years). The various decay chains are depicted in figure 3.

The isotopes lead-214, lead-212 and lead-211 are rare in nature due to their short half-life, and therefore scarcely discharged to the environment. Hence these short-lived isotopes are not considered here, although in case of the discharge of a radionuclide like radon-222, these isotopes may contribute to the total radiation dose [Ti92]. The radioactive isotope $^{210}$Pb has a half-life of 22.26 years and is part of the decay chain of uranium-238 (see figure 3). Since uranium-238 is widely distributed in nature, the isotope $^{210}$Pb is also abundant in nature. The concentrations of the radioactive isotope $^{210}$Pb in lead is on the order of 1 Bq $^{210}$Pb per gram lead, i.e. only 0.1 pg $^{210}$Pb per gram lead [Ja69].

Decay of $^{210}$Pb results in the radioactive isotopes $^{210}$Bi and $^{210}$Po (see figure 3) with a half-life of 5 and 138.38 days, respectively. Since these are radioactive substances, $^{210}$Bi and $^{210}$Po are carcinogenic. Therefore, in an assessment of the consequences of the emission of $^{210}$Pb to the environment, the formation of $^{210}$Bi and $^{210}$Po has to be taken into account.

Lead is often present as a contaminant of natural ore, along with the radionuclides $^{226}$Ra and $^{210}$Pb. Industries using phosphate rock (containing relatively high concentrations of
radionuclides of the U-238 chain) emit large amounts of $^{210}$Pb. Industries using the wet process (e.g. phosphate fertilizer industries) emit watersoluble phosphogypsum containing large amounts of $^{210}$Pb into the water. The thermo process (e.g. used by brick- and China producing industries) results in the emission of $^{210}$Pb in the off-gases. Furthermore, the discharge of $^{226}$Ra and $^{222}$Rn results in a diffuse source of $^{210}$Pb in the environment. An overview of the emissions to air and water of some important industrial sources of $^{210}$Pb is given in table 1.
Table 1  Overview of some industrial sources of lead (estimated emissions in the Netherlands in GBq per year, [Bl91])

<table>
<thead>
<tr>
<th>Emissions</th>
<th>water</th>
<th>air</th>
</tr>
</thead>
<tbody>
<tr>
<td>coal fired power plants</td>
<td>-</td>
<td>6</td>
</tr>
<tr>
<td>thermal process phosphate industry</td>
<td>50</td>
<td>150</td>
</tr>
<tr>
<td>wet process phosphate industry</td>
<td>700</td>
<td>-</td>
</tr>
<tr>
<td>steel producing industry</td>
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<td>100</td>
</tr>
<tr>
<td>brick producing industry</td>
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<td>150-290</td>
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<tr>
<td>China producing industry</td>
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<td>&lt; 190</td>
</tr>
<tr>
<td>cement producing industry</td>
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<td>&lt; 70</td>
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<tr>
<td>remaining</td>
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<td>&lt; 100</td>
</tr>
<tr>
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<td>400-900</td>
</tr>
</tbody>
</table>

3.3 Transfer and exposure pathways

Following the discharge of lead compounds into the environment, humans and ecosystems are exposed via various exposure pathways. The relative importance of the exposure pathways depends on diverse processes. In order to assess the effects and risks to humans and ecosystems a model of the entire source-to-effect chain is required, taking into account all relevant processes and exposure pathways. As an illustration of the demands on such a model a simplified overview of the source-to-effect chain is given in this chapter. Since several studies are already devoted to this subject [see e.g. Wh90, Bl92], only the most important processes are briefly summarized. An overview of the source-to-effect chain is given in figure 4.

3.3.1 Emission

As indicated in chapter 3.2, the lead emission into the environment takes place in the atmosphere, surface waters and the soil. As a result of differences in the emission sources and the exposure pathways, it is useful to subdivide the emission to the soil in an emission to agricultural land and an emission to non-agricultural land. In order to calculate the consequences of the discharges, several characteristics of the emission have to be known, including:

- the location
- the quantity of the discharged lead as a function of time
Figure 4  Overview of the most important exposure pathways for lead and $^{210}$Pb

- the chemical species of the discharged lead
- the particle size distribution

3.3.2  Dispersion and chemical conversion

Once the characteristics of the emission are known, the dispersion and chemical appearance of the lead in the environment have to be determined. Several processes can be identified affecting the dispersion and chemical appearance of lead in the environment:

- transfer between the atmosphere and the soil (deposition, resuspension)
- transfer between the atmosphere and the surface water (deposition, water spray)
- transfer between the surface water and the soil (flooding of a river along with sedimentation, dredging, groundwater discharge, run-off)
- atmospheric dispersion and chemical conversion in the atmosphere
- dispersion in surface waters, sedimentation and chemical conversion
- dispersion (both by physical and biological processes) in the soil, chemical conversion (dissolution and accumulation), infiltration to ground water and irrigation with ground water

The properties of the various processes listed above, depend largely on the chemical characteristics of the emitted lead compounds and on the properties of the medium in which the dispersion is considered. For example, the decomposition of organic lead in the
atmosphere depends on the solar flux; the transport and/or accumulation of lead in surface water, sediments and the soil depends on parameters such as pH, redox potential, microbial activity, etc.

3.3.3 Exposure pathways

The three main inputs of lead into the human body are drinking water, food consumption and inhalation [Kr90]. On average the figures for lead are as follows:

1. Approximately 35% of the total body burden can be attributed to drinking water. Drinking water is processed from surface water or groundwater. The use of lead pipes in the drinking water supply of some areas gives a substantial contribution to the total body burden.

2. An estimated 45% of the total body burden comes from food intake. Contamination of meat and dairy-products results from lead in drinking water, forage and soil, ingested by livestock. The contamination of crops occurs both by direct deposition (wet and dry deposition from the air as well as deposition due to irrigation with contaminated groundwater) and by root uptake. However, several other ingestion pathways can be identified, such as the consumption of fish and the intake of soil by children.

3. 20% of the total body burden stems from the inhalation of airborne lead and resuspended soil particles.

Due to the differences in the sources of emission, the relative contribution to the total body burden of the important exposure pathways differ for $^{210}\text{Pb}$ from the relative contribution of the important exposure pathways for lead. Furthermore, in case the consequences of the emission of $^{210}\text{Pb}$ are to be considered, external radiation has also to be taken into account.

In case of atmospheric emissions of $^{210}\text{Pb}$ into the environment the most important pathways are inhalation, external irradiation, deposition on open water or on agricultural lands followed by consumption of derived produce as mussels, milk and meat, respectively.

When contaminated river sludge is dredged and disposed on land, terrestrial pathways like the contamination of meat and milk of livestock by $^{210}\text{Pb}$ play an important role.

Processes relevant to the contamination of crops and live stock are considered, to a large extent, identical to the processes relevant to the contamination of flora and fauna, respectively. Therefore the contamination of flora and fauna is not considered explicitly. However, the accumulation of lead along the food chain has to be considered carefully.
3.4 Effects of lead and $^{209}$Pb

3.4.1 Effects of lead in humans

The health effects of lead in humans strongly depend on the chemical and physical form in which it occurs: metallic, organic or inorganic, and radioactive or not.

Metallic lead (as emitted through hunting, fishing etc) is chemically very stable and inert in the environment. Since in this form it is not absorbed in biological tissue, it has almost no direct effect on the health of humans, animals and plants. Through dissolution it can become reactive in tissue. Ingestion of metallic lead (grains) can therefore lead to negative effects and can become lethal, e.g. to geese and ducks, which swallow grains while grazing.

Organic lead (alkyllead, mainly as tetraethyllead and tetramethyllead) is used as anti-knock in petrol. These very volatile chemicals are quickly absorbed in several organs (with brain tissue as an important sensitive receptor).

Because of the rapid excretion of alkyllead, the lead concentration in urine is a good indicator for exposure to organic lead and is given in mg Pb/l urine. In humans the normal background level of lead in urine is estimated to be 0.06 mg/l. In workers moderately exposed to alkyllead this can be 0.09 - 0.15 mg/l. When the Pb-concentration in the urine exceeds 0.15 mg/l some mild symptoms can be seen [Ja91]. The exposed groups are relatively small but the effects can be serious. After inhalation of organic lead these symptoms include fatigue, muscular pain, irritability, and abdominal pain. In some working conditions (working at petrol stations, with batteries and petrol) it can lead to intoxication within a period of several hours to days and cause vomiting, diarrhoea, headache, delirium and coma. Intoxication with alkyllead normally occurs at Pb-urine levels exceeding 0.3 mg/l [Ts86].

In the human body inorganic lead has no biological essential function but it interferes with different enzyme systems. It accumulates in the body and inactivates enzymes, destroys the structure of cells and interferes with the cellular metabolism [JEFCA86]. Exposure to low doses can have far-reaching effects on essential enzymatic- and energy-transfer mechanisms. This can lead to distortion of the haem synthesis and affect the nervous system, the kidneys, the vitamin D synthesis and growth.

The retention of inorganic lead that enters the body via inhalation varies from 20-60%, depending on the aerosol size. The retention of ingested inorganic lead (from food and water) is 10% for adults, and between 40-50% for children. The biological half life ($T_{1/2,biol}$) is 36-40 days in soft tissue and blood, and $10^4$ days (± 27 yr) in bone structures. The absorbed lead is primarily deposited in bone structures (95% in adults and 70% in
70% in children). About 1% of the total body burden can be found in blood (mainly in the red blood cells).

Lead that is actually absorbed into the body is excreted mainly in the urine (about 75%) and further in faeces (about 15%) and in sweat, hair nails and bile (about 10%).

The concentration of lead in blood (PbB-value) is found to be a good indicator of the total body burden and is given in µg/l. Figure 5 gives an overview of the quantitative and qualitative effects of inorganic lead (in terms of NOEL and LOEL\(^1\) and of PbB-values).

At relatively low concentrations of lead in the air (as occurs in the Netherlands) the uptake of lead by children is approximately double the amount of lead uptake by adults [WHO87, GR84, Kr90]. Therefore, there is general consensus that the most vulnerable group is those of young children and infants (0-6 yr) [GR84]. The main reasons are:

- young children and infants ingest more lead from dust, soil and dirt via their hands than adults,
- the food uptake from infants and young children per kilogram bodyweight is higher; also the fractional resorption from the intestine is higher (up to 40-50% instead of 5-10% in adults),
- the transport of lead to the vulnerable organs is higher (other "kinetics"): the blood-brain barrier is more permeable for lead, and the uptake in bone structures is relatively smaller than in adults,
- the vulnerability of the organs (esp. blood and nervous system) in children and infants is higher than in adults.

Because of the low probability of exposure to high concentrations of organic and metallic lead, these forms are not a serious health threat for the general public. Therefore we will concentrate on the effects of inorganic lead, and from now on in this report the term 'lead' will be used for 'inorganic lead' unless stated otherwise.

The biological effects of radioactive \(^{210}\text{Pb}\) whether metallic, organic or inorganic, are caused by the radioactive decay of the \(^{210}\text{Pb}\) and of the daughter nuclides in the body. The largest contribution to the radiation dose following the intake of \(^{210}\text{Pb}\) is mainly of the daughter nuclide \(^{210}\text{Po}\), being an α-emitter.

Exposure to ionizing radiation can cause both deterministic effects and stochastic effects. Deterministic effects only occur above a certain threshold. As the radiation dose following exposure to \(^{210}\text{Pb}\) is usually well below the lowest threshold value for an organ of the body, deterministic effects are not considered here.

Stochastic effects are effects without a threshold value. The probability of the effect increases with increasing exposure. For exposure to low doses of radiation a linear

\(^1\) NOEL = No Observed Effect Level
LOEL = Lowest Observed Effect Level
relationship between the radiation dose and the effect is assumed. Several stochastic effects are distinguished: morbidity as a result of cancer, mortality due to cancer, damage to the health of an unborn child and genetic effects. Each type of effects has its own dose to risk relationship [ICRP91].

![Haematologic symptoms](image)

**Haematologic symptoms**
- Inhibition of ALAD in erythrocytes
- Increased concentration of protoporphyrin in ery’s
- Increased excretion of ALAD in urine
- Shorter life of erythrocytes
- Reticulosis
- Anaemia

**Other symptoms**
- Subjective symptoms
- Effect on the central nervous system
- Effect on the peripheral nervous system
- Colic
- Effect on the kidney function

![Figure 5](image)

Figure 5 Relation between the concentration of lead in blood (PbB-levels) and the appearance of haematological and other symptoms in humans [Kr90].

Ingested lead is primarily deposited in bone, liver and kidneys. Since the biological half life is considerably larger in bone structures than in soft tissue, the most sensitive receptor for $^{210}$Pb is the bone surface.

### 3.4.2 Effects of lead in animals and ecosystems

Lead can have various effects on terrestrial and aquatic organisms. Janus *et al.* give an extended review of publications on this subject [Ja91].

Many of these studies are mainly based on the parameters growth, reproduction, development and survival. Effects on behavioral and physiological processes are not included, because their influence on the existence of the species is unknown. For many species one or more NOEC-levels referring to lethal or sublethal effects are available. It is evident that the NOEC’s for different effects and different species can vary greatly. From these NOEC’s the maximum permissible concentration (MPC) is calculated, the

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1. NOEC = No Observed Effect Concentration
maximum concentration of a pollutant in water or soil at which no unacceptable effects on the ecosystem are expected.

For deriving toxicological recommended values for the protection of ecosystems 'single-species' NOEC's are used. Different methods are developed. In a comparative study by the Health Council of the Netherlands five of these methods have been compared to each other [GR88]. Although none of these methods can pretend to protect the ecosystems from any negative effect of toxic substances, they form a good starting point in using the present knowledge for the protection of ecosystems. At the RIVM a modification of the Van Straalen method is used [Ja91]. With this modification the '95%-protection level' can be calculated, based on the single-species NOEC-values. This is the concentration at which, theoretically, 95% of the species present in a given ecosystem or compartment is protected (for 95% of the species the NOEC is higher than this concentration, and for the remaining 5% negative effects might be expected).

In aquatic ecosystems negative effects leading to reduced growth can be found at levels as low as 15 mg/m³ (in trout, Salmo gairdneri). In fish the haem synthesis can be affected at concentration of 2 mg/m³ and higher [Kr90].

At a lead concentration of 19 mg/m³ the watersnail Lymnaea palustris dies and mollusca disappear, so sublethal effects must appear at much lower levels. Concentrations that are even 50 times lower result in strong absorption in zoo- and phytoplankton (up to 465 mg/kg). This can lead to accumulation of Pb in the foodchain and therefore to negative effects in animals higher in the foodchain.

In terrestrial ecosystems at Pb concentrations exceeding 200 mg/kg soil the microflora as well as the fauna are affected. The biodiversity (esp. of nematoda and enchytraea) and the total biomass is reduced. Reproduction also decreases. Decreased pH (acidification of the soil) reinforces these effects. Lead grains from hunting will primarily affect the fauna. From studies concerning ducks, geese, pigeons and eagles it is known that even the ingestion of one or two grains can be lethal [Kr90]. Root-uptake of lead by vegetation does not affect its concentration in the above-ground parts. This concentration is, however, affected by leaf uptake from dry as well as wet deposition. This can be especially important in mosses, where high accumulation of lead can, upon consumption by fauna, be brought into the foodchain.

Since the bioavailability of lead in the soil depends on the properties of the soil, it is not possible to define a single MPC for the soil. Reference values for lead in the soil are therefore defined as a function of the content of organic material and clay in the soil [Ja91].

The starting point of the protection of ecosystems in environmental radiation protection
policy is the perception that individual animals and plants are in general less susceptible to radiation than human beings [VROM91]. Since the protection of an ecosystem is on population level, and since exposure to radiation levels higher than the average exposure of humans is likely to occur only on a very local scale, it is argued that the protection of human beings to radiation embodies the protection of the ecosystems. Hence there are no environmental Maximum Permissible Concentrations for radionuclides defined for ecosystems.
4 PRESENT SITUATION WITH RESPECT TO THE AVAILABILITY OF MODELS

4.1 Introduction

Models vary largely in complexity and range, depending on the application of the model. In general, two different types of integrated models can be distinguished, as indicated in chapter 2.

An integral model describes the entire chain from source-to-effect (and risk) in a simplified manner. In this type of model the results of experiments and the results of calculations with complicated models are often used as simple relationships. Since these models require relatively little computing time they are used to calculate a vast number of scenarios for one or several policy themes.

An integrated model also describes the entire chain from source-to-effect (and risk). However, this type of model usually consists of linked complicated submodels. A submodel, which is mostly physical, chemical or biological process oriented, describes a small part of the source-to-risk chain, such as the dispersion of substances in the atmosphere, the chemical speciation in the soil or dose effect relationships. Since these models are complex, they require relatively large computing times and their application is usually limited to a single scenario and a single source.

Both types of models are currently in use at the RIVM. In this chapter a preliminary overview is given, based on the catalogue of environmental models at the RIVM [Br92] and interviews with experts of the RIVM. A pre-selection is made according to the expected applicability of the models to the pollutant lead. For more detailed information on the listed models, the reader is referred to the catalogue of environmental models and the included references.

4.2 Models covering the entire source-to-effect chain

Integral models are currently in use at the RIVM to assess the expected state of the environment, resulting from a variety of policy scenarios.

An example is the model system EXPECT. It is used to assess the environmental and economic impact of environmental policy scenarios. EXPECT describes the flow of pollutants in the environment from emissions to effects on ecosystems and humans. The time step used in the calculations is one year and the output of the model consists of economic, ecological and environmental indicators.

In order to assess the risk and hazard of the use of chemical substances two models are under development at the RIVM, named PBS (Prioriteitssysteem Bestaande Stoffen, priority assessment system for existing (organic) substances) and BNS (Beoordelingssysteem Nieuwe Stoffen, risk assessment system for new (organic) substances). These steady state models describe the chain from emission to environmental
distribution. Several processes are implemented via the use of other models, like OPS, SimpleBox and SimpleTreat. Information on the kind of effects, however, is not given.

The BNS system can be used for local scale calculations. The outcome consists of risk quotients (the ratio of the Predicted Environmental Concentration and the No Effect Concentration, PEC/NEC) for ecosystems and for humans, based on average exposure.

The PBS system can be used for calculations on a local scale as well as on a regional and national scale. The PBS system ranks substances from harmful to relatively harmless, based on some worst-case assumptions. Although the structure of PBS seems to be adequate, it can not (yet) be used for lead. A significant effort to implement all the necessary exposure pathways and parameters for lead would be needed.

There are some (simplified) models covering the entire source-to-effect chain. The environmental compartment models BIOS and MiniBIOS describe the (time dependent) distribution of radionuclides in a model biosphere, composed of aquatic and soil compartments. The resulting radiation dose to humans is calculated.

The distribution of micropollutants in the environment can be calculated with the environmental compartment models SimpleRisk, SimpleSal and SimpleTreat. The output of the model consists of steady state equilibrium values for the concentrations in the boxes (air, water, sediment, agricultural soil, natural soil, suspended solids, biota). However, in general these models are too simplified to be used in a risk assessment for lead. These models are therefore not included in the overview in chapter 5.

4.3 Models covering part of the source-to-effect chain

Integrated models can be made by linking complex independent compartment models. The output of one model is used as the input of the next model. Recurrences may also be introduced in the system. The 'sub'models describe parts of the complete chain from source to effect (to risk) and are usually limited to the processes in a single compartment (e.g. atmosphere, surface water, soil).

4.3.1 Emission

There are several models available to determine the emission of a source. In these models the knowledge of an industrial process is used to calculate the emission per unit production. An example of such a model is MODEL METAL FINISHING, a model to assess the amounts of the most important components in the effluent, sludge and concentrated solutions of a model metal finishing shop. It is clear that application of this model is limited to only a few sources.

More generally applicable are the system RIM+ and the STRAVERA model (linked to RIM+, but specifically designed for calculations of total emissions of pollutants to surface waters) to determine the emissions of processes. RIM+ consists of a database with
information on emission sources, i.e. the processes (e.g. industrial, transport), on technical measures, volume measures and the like. The emissions of a particular source are calculated by multiplication of the volume of a process (e.g. production in tons/year) with an emission factor (e.g. emission of CO₂ per unit production), based on technical specifications of the process. Information on the discharge of lead compounds has not yet been introduced in the RIM+ database. However, the structure of the database is appropriate for storage of lead emission data and calculation of emission scenarios.

4.3.2  Transport, dispersion and chemical conversion

Atmosphere
A number of models are currently available at the RIVM to compute the transport and dispersion of substances in the atmosphere. The models differ both in the geographical scale of application (local, regional or continental) and the time scale of application (either time averages or real-time calculations). Chemical conversion of substances is often modelled as a degradation, resulting in a depletion of the amount of polluting substance in the atmosphere. However, for some specific substances, such as NOₓ, models are available to assess the dispersion of secondarily formed components. The input of the atmospheric models consists of emission characteristics such as quantity, emission height, particle distribution and of meteorological data, while the output usually consists of the concentration in the air and the deposition on a grid. Deposition patterns can be used as input for models covering the dispersion in the soil and surface waters. Available models are:

CAR A model to calculate the dispersion in an urban environment on a small scale. The model is used to assess the (yearly averaged) concentrations of pollutants emitted by traffic.

OPS A model to compute the transport, dispersion and degradation of substances in the atmosphere on a local to regional scale. The model is used to calculate the (time period averaged) concentrations and deposition patterns of pollutants.

TREND A model, based on the same mathematical description of the processes as OPS, especially adjusted to acidifying components. The chemical transformation of the acidifying compounds and the transport of the secondary formed components are taken into account. However, compared to OPS the program TREND is less documented and more difficult to use.

EUPUFF A model to compute the transport, dispersion and degradation of substances in the atmosphere on a continental scale. The model is used to assess the (real time) concentrations and deposition patterns of pollutants.

For the emission source 'lead in petrol' the model CAR can be used to calculate the
emission and dispersion of lead on a very local scale near the motor road. The model TREND is already used at the RIVM to describe the dispersion of lead in air on a local and regional scale [Ba91]. The processes in the model OPS for the dispersion of lead are described in the same manner as in the model TREND. Therefore OPS can be used as well.

In OPS the growth and dispersion of the radioactive daughter nuclides $^{210}\text{Bi}$ and $^{210}\text{Po}$ is not modelled. However, as the maximum radiation dose occurs near the source, the transport time of $^{210}\text{Pb}$ in the atmosphere is limited to only a few hours. Following the emission of 1 Bq $^{210}\text{Pb}$, in one hour only a fraction of $4 \times 10^{-6}$ $^{210}\text{Pb}$ has decayed, resulting in approx. $6 \times 10^{-3}$ Bq $^{210}\text{Bi}$ and $6 \times 10^{-7}$ Bq $^{210}\text{Po}$. As the radiation dose due to unit intake is comparable the decay of $^{210}\text{Pb}$ and the growth of $^{210}\text{Bi}$ and $^{210}\text{Po}$ is small on this time scale, and the model OPS is also applicable to determine the dispersion of $^{210}\text{Pb}$ in air on a local scale.

**Surface water**

Relatively few models are available to compute the transport and dispersion of substances in surface waters. The few models available are either restricted to isolated aquatic systems (e.g. a single lake with sediments, like the Bilth(L) model) and are relatively simple. Available models are:

- **DILMOD**: A model to estimate the dilution of pollutants in effluents from waste water treatment plants discharged in surface waters. The model calculates the concentrations at a certain longitudinal distance downstream of the outlet with the use of dilution factors based on flow rates. The output of the model consists of concentrations in river water and river sediments, expressed in PEC/NEC ratios (Predicted Environmental Concentration/ No Effect Concentrations). The model DILMOD is suited for a simple assessment of the consequences on a local scale near the source.

- **BILTH (L)**: A simple spreadsheet model for the release of radionuclides in a single lake system, consisting of water, sediment and fish. Output data are time series of concentrations in the diverse elements of the lake system.

- **OKLSO**: A spreadsheet model that describes the distribution of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in surface water and sediment of the Scheldt estuary. The output data consist of place-related concentrations of both nuclides.

It is clear that the models listed here are not suited to determine the dispersion of lead in the aquatic environment on a regional or national scale. Furthermore, the listed models have been used only for specific applications and are up to now not used for lead. In case a more complicated approach to water transport and dispersion of pollutants in surface waters and sediments is required, the user is referred to various model systems, available outside the RIVM (see for example [Oe92]).
In the study "Second Environmental Outlook of the Netherlands" the model systems PAWN and MANS have been used for the aquatic environment [Ba91].

The model system PAWN (Policy Analysis of water Management for the Netherlands) was developed in the period 1976-1979 to support the decision making process in water management [Sa89]. The system consists of a distribution model, describing the distribution of water in the Netherlands, and a model for the flow of substances, describing the emissions and the processes in the surface water systems. The PAWN system is expanded and updated by the institute RIZA (National Institute for Inland Water Management and Waste Water Treatment). For North Sea management purposes, a simplified PAWN model is presently being developed in the MANS (MAnagement of the North Sea) project.

In the near future the model CHARON of the institute "Delft Hydraulics" will be operational at the RIVM. CHARON models the transport and chemical speciation of substances in both the soil, ground water and surface waters. Furthermore, modules are available to assess the resulting concentrations in the aquatic food chain. For a description of the model CHARON the reader is referred to [Ro91].

Soil and groundwater
Several models are available to describe the accumulation and transport of pollutants in the compartment soil and groundwater. However, two distinct types of models can be identified.

The convective and dispersive transport of pollutants in the water phase (groundwater) is described by models for a groundwater system, like FLORAN, AQ, and METROPOL. However, the chemical transposition of pollutants is described in these models in a very simplified way: only linear sorption and degradation in a water saturated system are taken into account. The models for the distribution of substances in the groundwater system are therefore not applied to lead.

The chemical transposition of substances in the non-saturated soil is governed by (non-linear) sorption, complexation by organic and inorganic ligands, precipitation and dissolution, redox reactions. The models EQ3/6, ECOSAT and MINEQL+ are used to evaluate the chemical speciation, determined by these processes. The transport of pollutants in the soil by e.g. bioturbation and ground water transport is not included in these models.

A model to simulate accumulation and transport of heavy metals in soil columns is under development as a special version of SOTRAS. A model describing both the chemical speciation and the transport of lead in ground water is not operational at the RIVM. Combining the model CHARON (as indicated in the section surface water), with one of the operational 2D or 3D ground water transport models may fill this gap.

The accumulation of heavy metals in agricultural soil and crops can be calculated with the model NMPCULTY, as used in the study "Second Environmental Outlook of the Netherlands" [Ba91].
4.3.3 **Intake and uptake of pollutants**

Once the concentrations of the substances in the different environmental compartments are known, the concentrations in flora, fauna and human food are usually calculated with the aid of concentration factors and transfer factors. However, there are some models available to describe the (time dependence of the) transfer of pollutants in livestock. Examples of these models are:

**BILTH (C)** The Bilth (cow) model describes the time dependent concentrations of radionuclides in milk and meat following air contamination.

**DCOW** The DCOW model describes the kinetics of 2,3,7,8-TCDD in cows.

The human intake of pollutants is usually calculated by the selection of a representative individual along with various assumptions with respect to the consumption pattern, the origin of the food and behaviour (like inhalation rate of air). An example of the various assumptions made on the consumption pattern of the individual, as considered in a safety assessment for the emission of radionuclides, is given in the MORIS study [BI92].

For lead the intake of the pollutant by ingestion and inhalation are the two main exposure pathways. For the radionuclide $^{210}\text{Pb}$, apart from ingestion, exposure to gamma-radiation due to the decay of radionuclides in the air and on the soil contributes to the radiation dose. Consequently a model is required to determine the radiation dose due to external radiation. An example of such a model is EXPO, describing external radiation from a contamination of the soil [La93].

4.3.4 **From dose to risk**

Once the intake of harmful substances is determined, a dose-effect relationship is required to determine the effects. Dose-effect relationships are either derived from observations of exposed groups and/or experiments on laboratory animals, or from metabolic models. The intake of substances and/or the concentration in the relevant organs are usually compared to parameters like NOEL, LOEL and LD50 (Lethal Dose 50%).

In radiation protection, models describing the human metabolism are frequently used to determine the risk to develop cancer, following the intake of radionuclides or exposure to external irradiation. Examples are the ICRP models, currently in use at the RIVM, to determine the effective equivalent dose (and consequently the risk to develop cancer) resulting from the intake of radionuclides [ICRP82, ICRP91]. These models take into account all the relevant exposure pathways in the human body (from ingestion as well as

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1 Intake: 'consumption' through food, water or inhalation  
Uptake: absorption into blood and tissue
inhalation), the organs involved and the effectiveness of the radiation. Final outcome is 
the effective equivalent dose \( H_{50} \) in units of Sievert, being the weighted effective dose for 
the next 50 years after exposure.

In these calculations also the effects of exposure to daughters and granddaughters are 
included. Two different models, one for inhalation and one for ingestion can be 
distinguished. In figure 6 a schematic overview is given for the inhalation and ingestion of 
\( ^{210}\text{Pb} \).

From the effective equivalent dose the mortality risk can be calculated. With a proposed 
mortality risk factor of 2.5\% per Sievert a mortality risk of \( 10^{-5} \) per year corresponds to 
an effective equivalent dose of 0.4 mSv per year [VROM91]. With the use of a dose 
conversion factor of \( 1.5 \times 10^{-6} \) Sv/Bq ingestion, the mortality risk limit of \( 10^{-5} \) per year is 
reached by the ingestion of 260 Bq \( ^{210}\text{Pb} \) per year, equivalent to only 0.1 ng \( ^{210}\text{Pb} \) per 
year [No85]. The yearly intake of lead is approximately 25 mg per year [Kr90], whereas 
the activity of lead is on the order of 1 Bq.g\(^{-1}\) [Ja69]. Hence we conclude that on one 
hand the contribution of radioactive lead to the total lead burden is negligible, on the other 
hand the contribution of the ingestion of lead to the radiation dose is also negligible.
Figure 6 The ICRP model for the ingestion and inhalation of lead-210.

F  = fraction transferred to the next compartment  
T  = half life time in days  
Ing = Ingestion;  
NP  = Nose-Pharynx  
TB  = Tracheo-branchial  
Inh  = Inhalation  
P  = Pulmones  
L  = Lymphé

4.3.5 Modelling an ecosystem

Models that predict the effects of lead (or heavy metals in general) on natural ecosystems were not found. Nevertheless, some of the models mentioned earlier have modules that give some information on ecosystem effects:

PBS and BNS  see paragraph 4.2
CATS  describes the accumulation (in both biotic and abiotic compartments) of cadmium in meadows, and can perhaps with some modifications be used for lead.
For the effects on ecosystems, following the methods used in PBS and BNS, the PEC/MTR value for a given compartment can be used to give an impression of the influence on ecosystems. When the PEC/MTR value is smaller than 1 (more than) 95% of the species in the ecosystem will not suffer significant negative effects from the lead burden. The smaller the PEC/MTR value, the safer the ecosystem is. When the PEC/MTR value is greater than 1, less than 95% of the species present in that (undisturbed) ecosystem will be protected from significant negative effects due to the total lead burden.
5 EVALUATION OF THE INTEGRATED CHAIN MODEL

5.1 The source-to-effect chain covered?

From interviews with modelling-experts from the laboratories involved it became clear that the approaches to the source-to-effect chain described here differ in several ways.

For some problems a source oriented approach is used, in which the effect of one single source (or a change in the release of a source) must be calculated. An example of this type of modelling is the dioxin model DCOW.

Other problems are solved with a more region oriented approach, in which all the sources (local as well as diffuse) within a defined region (e.g. the Netherlands) are considered for calculating the total effect of a chemical substance.

This difference in approach is probably caused by the fact that the evaluation of the environmental quality in some cases is source-oriented with demands in terms of effects or additional risks per source, whereas in the other cases the demands are more in terms of a acceptable concentration at a given location, which of course is the result of all the sources together. This discrepancy in approach is already mentioned earlier in the AIM study [Ba91].

Consequently, several different spatial levels have to be distinguished. In the figures 7, 8 and 9 the availability of models on three different spatial levels is presented. The three levels are local (figure 7), regional (figure 8) and national (figure 9). In boxes the different compartments or subcompartments are given in which specific physical and chemical processes (can) occur. The arrows represent processes that characterize the transformation to other (sub)compartments.

For processes that occur on a short time scale (in the order of hours) toxic and radioactive lead can for most applications be described with the same models, as the half life of $^{210}\text{Pb}$ is 22 years. For long time scale processes other models that include radioactive decay and ingrowth of nuclides are necessary. This implies that the atmospheric dispersion and the human exposure can be modelled with the same models, but for the accumulation and other (transfer- or chemical and physical) processes in soil and vegetation other, pathway specific models are needed.

Describing the ecosystem value is not based on the effects on the most sensitive species, nor is it based on the most valuable species. The choice for specific species is in most cases motivated by their availability and suitability for experimental purposes.

Nevertheless, based on the single-species data and with extrapolation methods like the one from Van Straalen et al one is able to pronounce upon the influence on the total ecosystem [GR88].
For the calculation of the total burden to humans, data on the food range (as described in MORIS) and ICRP-models (for the radioactive dose from ingestion and inhalation) can be used. The resulting health effects and mortality risks can be calculated using the available NOEL’s and the ICRP-models.

From the figures it becomes clear that for the description of the dispersion of lead and the total human burden on a local scale a relatively large number of models is available, although some pathways are not covered (see figure 7):

- distribution from water to terrestrial plants and animals (uptake), and to aquatic plants
- distribution form aquatic plants to sediment (biodegradation) and vice versa (uptake) and the external radiation from water and sediment to humans

Many of the remaining processes are only partly modelled on different levels of
complexity. Although the diagrams suggest that certain pathways are covered by a specific model, some of the models only give a description of parts of the pathway. For instance Bilth(C) calculates the radiation dose from the consumption of milk (and milk products) and beef. It does not cover the total radiation dose from terrestrial animals, nor does it cover the total dose from meat consumption because in the Netherlands the beef consumption is less than half the total meat consumption.

Although Bilth(C) and Bilth(L) are developed especially for radionuclides they can also be used for toxic substances. There are also models which have not specifically been developed for lead, but can be used for lead as soon as the relevant parameters for lead are available. RIM+ is an example of this.

Exposure from aquatic animals (fish and fish-products, mussels, etc) can be important for some groups of the population that are extreme consumers of these foodproducts. When such a specific risk group is to be taken into account the food pattern has to be adapted. In the MORIS report and in the booklet "Wat eet Nederland" guidelines are given [BL92, WVC88].

Because the data and parameter values are based on average values of consumption,
soil type, weather etc, they lead to an average individual dose in humans.

Comparing the figures 7, 8 and 9, it becomes clear that the larger the scale to be modelled, the smaller the availability of models. Models for several pathways are missing, and it is not expected that they will be available in short time. Perhaps the extension or adaptation of existing models like PBS and EXPECT for lead can give the possibility to fill the gaps in a relatively short time. On the other hand integrated models like BIOS can be used, although one must realise that they use extensive simplifications of pathways and intercompartment relationships.

5.2 Linking the available models

The various compartment models can be linked off-line or on-line. On-line linking implies that multiple models have to be run simultaneously, while exchanging information. On-line linking is complicated. It is required if the individual models have a two-sided feedback, within the time step of model calculations. If the output of one model can be considered as a boundary condition for another compartment model, off-line linking is acceptable. A first step towards off-line linking of various compartment models is
standardization of formats of input and output data. At the RIVM a first modest step towards standardization of model input and output data is made through the introduction of quality assurance systems for models. This introduction includes among other activities the writing of Standard Operation Procedures (SOP) for the available models. The SOPs should include an extensive description or a reference to such a description of all input and output data. Presently SOP's for computer models are prepared at the RIVM and related institutes. A catalogue of RIVM models, briefly summarizing also input-output characteristics, was published in 1992 [Br92]. A next step toward off-line linking would be to standardize the identification codes, the units and the write-formats of input and output data. To implement this second step for all relevant RIVM models requires a considerable investment in time.

A fundamental problem could arise when temporal and/or spatial scales of models would differ. Down-scaling of output of one model, to meet a requirement for higher spatial or temporal resolution of the next model in the chain, will increase the uncertainty in the results. In view of the large effort and fundamental problems related to an overall standardization of model data for off-line linking, off-line linking should be limited to specific chains, for which there is an immediate need. As individual chain models are generally developed for independent application, the system boundaries will be chosen in such a way that boundary conditions can be quantified. This means that compartment models often contain lumped descriptions of adjacent compartments, in order to enable quantification of boundary conditions. Before off-line linking of two independently developed compartment models, it should therefore be considered whether the results of the coupled model are better than or at least compatible with the original stand-alone model which includes a lumped description of the preceding compartment in the chain.
6 CONCLUSIONS AND RECOMMENDATIONS

The pilot project to study the possibility of constructing an integrated chain of models for lead and $^{210}\rm{Pb}$ has once again documented two different approaches for modelling the source-to-effect chain used at the RIVM.

Integral models such as the EXPECT system have been developed specifically for the production of the Environmental Outlook reports. Model development within the context of projects is one of the recent organizational developments in RIVM that will be driven by definitions of the output variables, a set of environmental quality indicators and environmental policy indicators: together, these constitute the framework of the Environmental Outlook reports. For these applications integral models are likely to be used. However, integral models cannot be used for all questions on policy and research, since these models are appropriate to generating answers at aggregated spatial and time scales. For questions demanding analysis of the chain at a high spatial and/or temporal resolution, integration of stand-alone deterministic models is still necessary. These integrated models are also needed to support the development and use of integral models.

Several difficulties arise in the integration of stand-alone compartment models. Surveying the models for lead and $^{210}\rm{Pb}$ shows that at the moment large gaps exist in the modelling of the complete source-to-effect chain for lead and $^{210}\rm{Pb}$. This is most evident on the regional and national scales. Although models to fill some of the gaps are currently in development, it has been shown that on these spatial scales achieving an integrated model of the source-to-effect chain with the present-day set of models is not yet possible. To cover the whole chain, new models either need to be developed in house or be acquired from other institutes.

On a local scale, several models for constructing an integrated chain of models are available, although differences in applicability and complexity are expected. Therefore it can be worthwhile in some projects to consider linking of these models off-line with the use of prescribed input/output formats. However, even when the models to construct an integrated chain of models are available, several difficulties may arise. The process of linking can be very time consuming, and one has to be careful with the differences in the temporal and/or spatial scales of the models. The effort required for this type of integration is still considerable and the result will remain sensitive to input-output discrepancies between individual models.

It is therefore recommended restricting the development of integrated chain models to specific research questions. Hence, a possible follow-up of this project might start with an inquiry into the questions expected and assessing the requirements for the models to deal with these questions. It should be decided if answers to these questions can be obtained
with "simplified" integral models or "complex" integrated models. In the case of an integrated model needing to be constructed, it is recommended surveying the models outside the RIVM to fill the gaps in modelling the chain. Only when appropriate models are not available inside or outside the RIVM, should a new model be developed.

These recommendations are just as valid for lead as for $^{210}\text{Pb}$ in modelling an integrated source-to-effect chain. However, the survey on pathways and models has indicated two differences in (modelling the) source-to-effect chain for lead and $^{210}\text{Pb}$. Firstly, the sources of lead are both point sources, like industrial plants and diffuse sources, like traffic, whereas the sources of $^{210}\text{Pb}$ are only point sources. Secondly, for lead Maximum Permissible Concentrations are used for the protection of both human beings and ecosystems, whereas in radiation protection radiation doses are calculated for the most exposed individual, implying that the protection of ecosystems is incorporated in the protection of human beings. Due to these differences in both sources and end-points, models should be used for lead on local, regional and national scales, while for $^{210}\text{Pb}$ the use of models should be restricted mostly to a local scale. However, the approach to establishing an integrated chain of models will be the same for lead as for $^{210}\text{Pb}$.
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