



Risk assessment of lead emissions from anthropogenic cycle

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Received 4 November 2014; accepted 23 January 2015

Abstract: The risk assessment right from the source of emissions can effectively guide the pollution control. A model was established, consisting of four part: source estimation, environmental fate analysis, exposure analysis and risk assessment. The human health risk, ecological risk and total risk of lead emissions were assessed. The factors were estimated to indicate the environmental decrease and exposure probability. Of all the 1887 t emissions in China in 2010 (quantified in the previous work), it is turned out 1.3 t reached human bodies (0.9 mg/ca), and 2.7 t reached the ecosystem. Lead mainly came from the Use stage for the source while lead causing risk mainly came from the Waste Management & Recycling and Production stages. As for chemical forms, PbO contributed most to the human health risk and PbSO₄ contributed most to the ecological risk. PbSO₄, PbO and Pb altogether contributed 71% to the total risk, indicating these three chemicals should be taken priority for the risk management.

Key words: lead; source; human health risk; ecological risk; total risk; life cycle

1 Introduction

Lead is one of the most abundant and toxic heavy metals in the environment [1]. There are both natural and anthropogenic sources for lead emissions, and the anthropogenic sources dominate the emissions [2]. According to study, more than 95% of the lead within the biosphere is of anthropogenic origin [3]. Although the lead abatement programs are provided in many developed countries, lead risk is still an important concern in the developing countries such as China [4]. Threaten imposed by lead pollution to human health and ecosystem still deserves our intensive attention. In this context, the risk assessment can be used to support the decision making in lead pollution management.

A wide variety of studies are already done on lead risk assessment, which can roughly be classified into the human health risk assessment and the ecological risk assessment. Human health risk assessment is defined as the process which estimates the likelihood of adverse health effects on humans who may be exposed to chemicals in contaminated environmental media [5]. Up to date, the human health risk assessment is generally determined by gauging the concentration in the

environment, applying the method recommended by US environmental protection agency (EPA) [6,7]. On the other hand, the ecological risk assessment evaluates the likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors [8]. For the ecological risk assessment, the method of hazard quotient is most widely applied [9–11]. All the present studies provide a clear way to measure lead risk. However, they fail to take the pollution sources into account, which is an integral part of risk formation. A study attempted to assess the risk of lead losses, but it lacks a detailed speciation as well as the analysis of environmental fate and exposure [12]. Thus, the information on which process (or life cycle stage) should be paid attention to, and which chemical forms should be taken priority is still missing. This kind of knowledge is significant in helping direct limitation of social consumption and governmental resources management.

In this study, a model to assess lead risk was established. The source emissions were estimated and the environmental fate and exposure were analyzed. The factors were applied to estimate the environmental decrease and exposure probability. Finally, the risk scores showing the levels of risk were calculated.

$$R = Q \cdot f_e \cdot f_x \cdot E \quad (1)$$

2 Methodology

2.1 Framework for lead risk assessment

The framework for lead risk assessment consists of four components: source estimation, environmental fate analysis, exposure analysis and risk assessment (Fig. 1). The source estimation refers to the quantification and speciation of emissions from anthropogenic cycle. The environmental fate analysis refers to the analysis of the move and transformation of chemicals in the environment. The exposure analysis estimates the likelihood of exposure to chemicals in contaminated environmental media. And the risk assessment includes three types of risk: the human health risk, the ecological risk and the total risk.

The risk of lead emissions (R) can be influenced by four factors:

where Q is the emission quantity from the source with the unit of t, f_e is the fate coefficient indicating the quantity decrease in the environment, f_x is the exposure coefficient indicating the likelihood of lead exposure, E is the effect factor showing the toxicity of lead emissions, represented by the unit risk score in the Indiana Relative Chemical Hazard (IRCH) ranking system [13]. The IRCH ranking system provides the unit human health risk score, unit ecological risk score and unit total risk score for various lead chemicals. As the scores in the IRCH ranking system were obtained from the sum of the points assigned, therefore they do not have any units. In this study, the risk was calculated by multiplying the unit score by the emission quantities with a unified unit of t (ton). For easy comparison, the outcomes of the calculation were defined as scores without units.

The total risk for lead emissions integrates both the

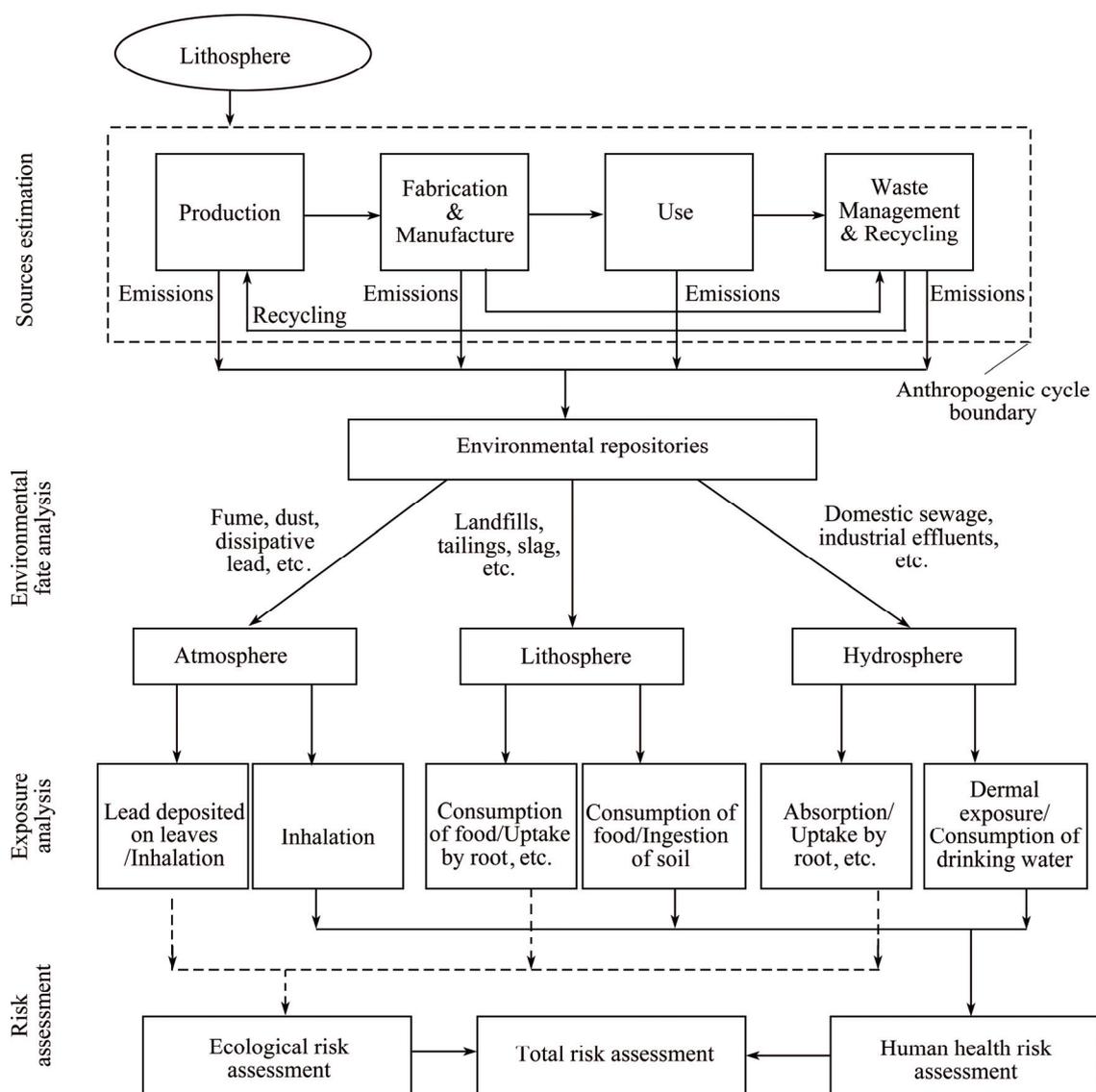


Fig. 1 Framework for lead risk assessment

human health risk and the ecological risk. According to the definition of total risk in the IRCH ranking system, the total risk R_T is defined as:

$$R_T = [(1.15R_H) + (R_E/3.5)]/2 \quad (2)$$

where R_H is the human health risk and R_E is the ecological risk. The coefficients of 1.15, 3.5 and 2 are applied by the IRCH ranking system based on how much the human health risk and the ecological risk contribute to the total risk.

2.2 Model for lead risk assessment

2.2.1 Source estimations

The anthropogenic cycle includes all the stages in lead life cycle: Production, Fabrication & Manufacture (F&M), Use and Waste Management & Recycling (WM&R). At the Production stage, the primary lead is refined from lead ores, and the secondary lead is refined from lead waste. At the F&M stage, lead product is manufactured. The Use stage is the phase where lead product provides service to human and satisfies the demands. At the Waste Management & Recycling stage, lead discards are recycled, disposed or landfilled. Due to the differences between the four stages, the lead chemical forms emitted vary from stage to stage. In our previous research, we have studied the quantities and chemical forms emitted from lead life cycle in 2010 for China, the detailed calculation can be referred to Refs. [14,15]. The chemical forms and their quantities are shown in Table 1.

2.2.2 Environmental fate analysis

In this section, what media the chemicals enter are firstly analyzed, and then the technological treatment and natural decrease are considered, which together result in a decrease of emissions quantity.

The environmental fate depends on the receiving media as well as the chemicals themselves. The types of lead wastes discharged from the life cycle stages have already been analyzed in the previous study [15]. In this study, the fate of chemical forms is estimated according to the possible fate of the wastes. For example, the tailings are emitted and mainly go to soil at the Production stage. As $PbSO_4$ mainly exists in the tailings, we assume that at the production stage, 80% of $PbSO_4$

goes into soil, and the rest goes into water and air with an equal proportion of 10%. The fate of all other chemicals can be obtained with similar analysis.

Due to the human treatment methods, some emissions are removed technologically when introduced into air, water and soil. According to China Statistical Report on Environment in 2010 [16], the removal rate from effluents by standard waste water treatment methods is 90%, with 10% still left in water. The removal rate of lead in soil is estimated to be 30% based on the treatment rate of multiple solid wastes recorded in the China Statistical Yearbook on Environment [17]. For the removal rate in the air, 95% of the total is assumed to be removed because the collection efficiency of dust collector in China is about 95% [18].

For lead emissions that stay in the environment, some have little mobility, and therefore they have minimal exposure potential and pose little or no danger to human and ecosystem. The decrease factors are used here to measure how much emissions exactly have the risk potential. For lead in the water, the precipitation reactions, along with strong absorption by suspended particle, cause a sharp decrease in quantity. The soluble lead is deduced from the solubility product constant [19]. For lead in the soil, lead is immobilized through sorption or precipitation processes, and usually remains on the surface. The soluble lead in soil is estimated from the absorption rate in soil [20]. For lead in the air, the atmospheric deposition, a transport process from air to water or soil, causes a quantitative decrease of lead in the atmosphere. This decrease factor is estimated based on the information that atmospheric deposition is about 1mg/d in polluted areas [21].

2.2.3 Exposure analysis

Exposure analysis includes the analysis of exposure endpoint, routes and likelihood. The endpoint of health risk is human body. The main routes for human exposure are the consumption of contaminated food and drinking water, the ingestion of soil and dust, the inhalation and dermal absorption. Lead in the soil can be exposed by food consumption or soil ingestion. For the food consumption, the exposure likelihood is represented by dietary absorption rate, which is 50% [22]. For the soil

Table 1 Chemical forms and their quantities emitted from anthropogenic lead cycle in 2010 in China

Life-cycle stage	Chemical form/t								
	PbO	PbSO ₄	Pb	PbCO ₃	PbO ₂	PbS	PbCl ₂	Others	Total
Production	66	160	0	110	6	89	0	18	450
Fabrication & Manufacture	47	5	19	0	0	0	0	1	72
Use	214	96	218	41	72	0	7	91	740
Waste Management & Recycling	195	176	70	45	69	2	41	27	625
Total	523	437	307	196	147	91	48	137	1887

ingestion, as the bioavailability of lead in soil is about 60% of that in water and food, therefore, the absorption rate of soil lead is 30% [23]. The inhalation and dermal intake are estimated based on the absorption coefficient [24] and the skin permeability [25], respectively. As for lead in the water, human can be exposed by water consumption or having bath. As a study on the drinking water in China showing that there is no health risk caused by lead [26], this work considers that the risk from drinking water is insignificant.

The endpoints of ecological risk include the animals and plants in ecosystem. Due to the richness of biodiversity and the complexity of ecosystem, the studies on ecological risk assessment just starts. The ecological exposure risk is related to many factors, such as body weight, contact route, and population dynamics. Some detailed exposure factors have been given for some specific animal species [27], however, a general factor has not been given for all the animals and plants. This work will not give the exposure factors for the ecological risk assessment, and just assume that all the lead emissions remaining in environment are to pose an ecological risk in the long run.

All the environmental decrease factors and exposure factors in the risk assessment model can be seen in Table 2.

2.3 Risk assessment

The risk scores were calculated for all three kinds of risks based on the unit risk scores from the IRCH ranking system [13]. The unit scores for chemicals such as PbSO_4 , PbS and Pb are already available from this ranking system, however, the scores for PbCO_3 , PbO_2 and PbO cannot be obtained. Estimations were made by applying the assignment principles of this ranking system, which indicates that the risk score of a chemical relies on its presence on the regulatory lists and target lists of the government.

According to the IRCH ranking system, the lowest ecological risk score is 75 for lead thiocyanate. Lead thiocyanate exists in the government regulation list of Dangerous for the Environment [28] while PbCO_3 does not. This indicates that PbCO_3 has a lower risk than lead thiocyanate according to the principals of this system. The PbCO_3 's risk score is assumed to be 74. As PbO and PbO_2 are on this very list, the score 76 is assigned to them.

Then, the unit human health risk scores for PbO , PbO_2 and PbCO_3 were estimated. Some information regarding the chemical's health effects was obtained from International Chemical Safety Card [29], using Pb as a reference. Pb , PbO and PbO_2 are similar in many properties except that PbO_2 is more active than Pb with strong oxidability. The unit risk scores of PbO and PbO_2

are estimated to be 22 and 23, respectively. As PbCO_3 has a low capacity to impact human health, its score is set as 11, using the scores for other metals in this system as the reference. All the unit risk scores for lead chemicals are shown in Table 3.

Table 2 Fate factors and exposure factors for lead emissions in risk assessment

Type	Subdivision	Factors	Reference
Decrease factors in environment	Decrease rate caused by water treatment	0.1	China Statistical Report on Environment [16]
	Decrease rate caused by solid waste treatment	0.3	China Statistical Yearbook on Environment [17]
	Decrease rate caused by dust collection	0.05	Ref. [18]
	Natural decrease factor in water	10^{-5}	Ref. [19]
	Natural decrease factor in soil	10^{-2}	Estimated from the absorption rate in soil [20]
	Natural decrease factor in air	10^{-2}	Estimated from atmospheric lead deposition of 1mg/d [21]
Exposure factors	Exposure by food and water consumption	0.5	Based on the absorption rate of food consumption [22]
	Exposure by soil or dust ingestion	0.3	Based on the lead bioavailability in vitro method [23]
	Exposure from inhalation	0.5	Based on the respiratory absorption coefficient [24]
	Exposure by dermal absorption	10^{-5}	Based on the skin permeability factor [25]

Table 3 Unit risk scores for lead chemicals according to IRCH ranking system

Chemicals	Unit human health risk score	Unit ecological risk score	Unit total risk score
Pb	22	140	33
PbCl_2	22	130	32
PbS	22	86	25
PbSO_4	31	100	33
PbCO_3	11	74	17
PbO_2	23	76	24
PbO	22	76	23
Others	22	103	29

3 Results and discussion

3.1 Human health risk assessment

Based on the model of lead risk assessment, the lead intake from food consumption was the most while the dermal intake was the least in 2010. It is known from the previous study that lead emissions were 1887 t from the source in 2010 for China, mainly from the Use stage (39%) [14]. However, after the processes of environmental fate and risk exposure, the lead emissions that reached human bodies were mainly from the Waste Management & Recycling stage (42% of the total) and the Production stage (33% of the total). The health risk also mainly came from the Waste Management & Recycling stage and the Production stage.

In terms of chemical forms, PbO (523 t) and PbSO₄ (437 t) were the most in the source emissions, occupying 28% and 23%, respectively [14]. After the processes of environmental fate and risk exposure, the quantity of lead emissions that reached human bodies was 1.3 t, which equaled 0.9 mg on per capital level, with PbSO₄, PbO and PbCO₃ were 0.29, 0.22 and 0.10 mg/ca, respectively. (the population in China was set as 1.36 billion). Based on Eq. (1) and the unit risk scores, the total human health risk score was calculated to be 30. For PbO and PbSO₄, their health risk scores were 12 and 7, respectively. Altogether, these two chemicals occupied 63% of the total. This fraction was higher than the quantitative fraction in the source emissions (51%). The distribution of human health risk among different life cycle stages and chemical forms are shown in Fig. 2.

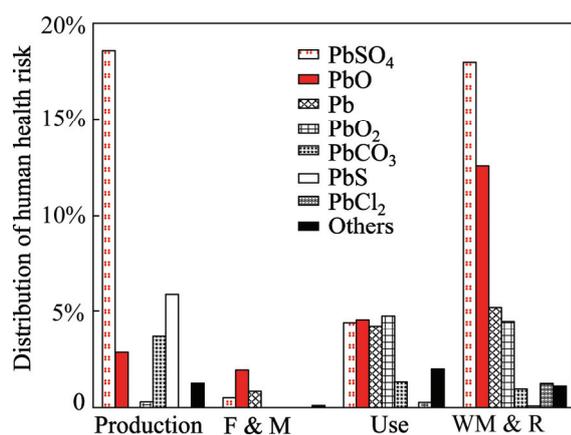


Fig. 2 Human health risk distribution for lead emissions in 2010 in China (F&M: Fabrication & Manufacture; WM&R: Waste Management & Recycling)

3.2 Ecological risk assessment

Following the assessment model, the lead emissions that reached the ecosystem were quantified as 2.7 t. For the source emissions, they mainly came from the Use

stage (as mentioned above), while the emissions posing an ecological risk mainly came from the Waste Management & Recycling stage and the Production stage (altogether occupied 65% of the total). Based on the unit ecological risk scores and Eq. (1), the ecological risk score was calculated to be 255, with the stages of Waste Management & Recycling and production contributing most.

In terms of chemical forms, the most chemicals emitted were PbO (523 t, 28%) and PbSO₄ (437 t, 23%). The most chemical quantitatively reached ecosystem was PbSO₄. For the total risk score, PbSO₄ (87 t, 38%), PbO (48 t, 28%) and Pb (42 t, 22%) contributed most. The distribution of ecological risk among the production and chemical forms are shown in Fig. 3.

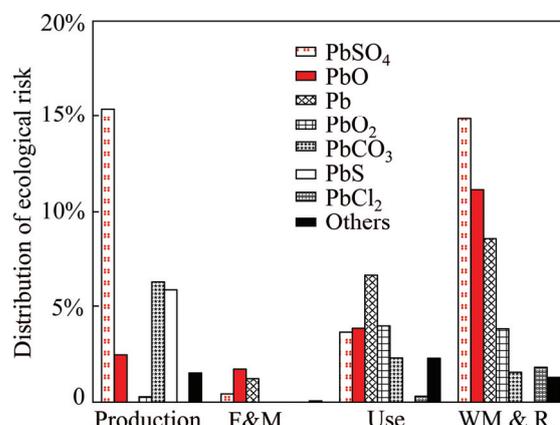


Fig. 3 Ecological risk distribution for lead emissions in 2010 in China

3.3 Total risk assessment

With Eq. (2), the total risk was calculated as 54. The total risk mainly came from the Waste Management & Recycling stage (43%) and the Production stage (32%). As for the chemical forms, the total risk was mainly caused by PbSO₄ (36%), PbO (20%) and Pb (14%). The ecological risk was larger than the human health risk, and the total risk score was between them (Table 4). The total lead risk score is significant in that the risk score of lead can be compared with the scores of other metals such as copper and zinc to measure their overall risks.

Table 4 Human health risk, ecological risk and total risk score in 2010 in China based on IRCH ranking system

Life cycle stage	Human health risk score	Ecological risk score	Total risk score
Production	10	80	17
Fabrication & Manufacture	1	8	2
Use	6	58	12
Waste Management & Recycling	13	109	23
Total	30	255	54

The quantity of emissions that caused an ecological risk was twice that caused the health risk, but the total ecological risk was over eight times the health risk, which probably resulted from the higher unit ecological risk scores (Fig. 4). For both risks, PbSO₄ and PbO were the primary chemicals that contributed most. Although the emission quantity of PbCO₃ causing risk was no less than that of Pb, its human and ecological risks were less. By applying Eq. (2), the chemicals' contribution to the total risk can be obtained from the chemicals' contribution to the human health risk and ecological risk. Pb contributed more than PbCO₃ to the total risk. To conclude, PbSO₄, PbO and Pb altogether occupied 71% of the total risk, which implies that these three chemicals should be taken priority for pollution control.

3.4 Uncertainty analysis

In this section, the main sources of uncertainty are identified:

1) The uncertainty of the hypothesis in the environmental process. When lead emissions entered the environment, their chemical forms will definitely change due to the interactions between them. In this study, the chemical forms from the source were applied to evaluate the risk, without considering the specific changes of chemicals in the environment, which may lead to some imprecision. In addition, the environmental fate of chemicals was estimated by assigning factors, which only showed a possible scenario in the environment.

2) Uncertainty in the exposure process. As the technology differs greatly from one area to another, the lead pollution is at different levels. In addition, some people are more vulnerable to lead pollution and thus are exposed to more risk.

3) Uncertainty in source estimation. The quantities of chemicals are quoted from the previous work, in which the chemical forms emitted perhaps experience one transformation.

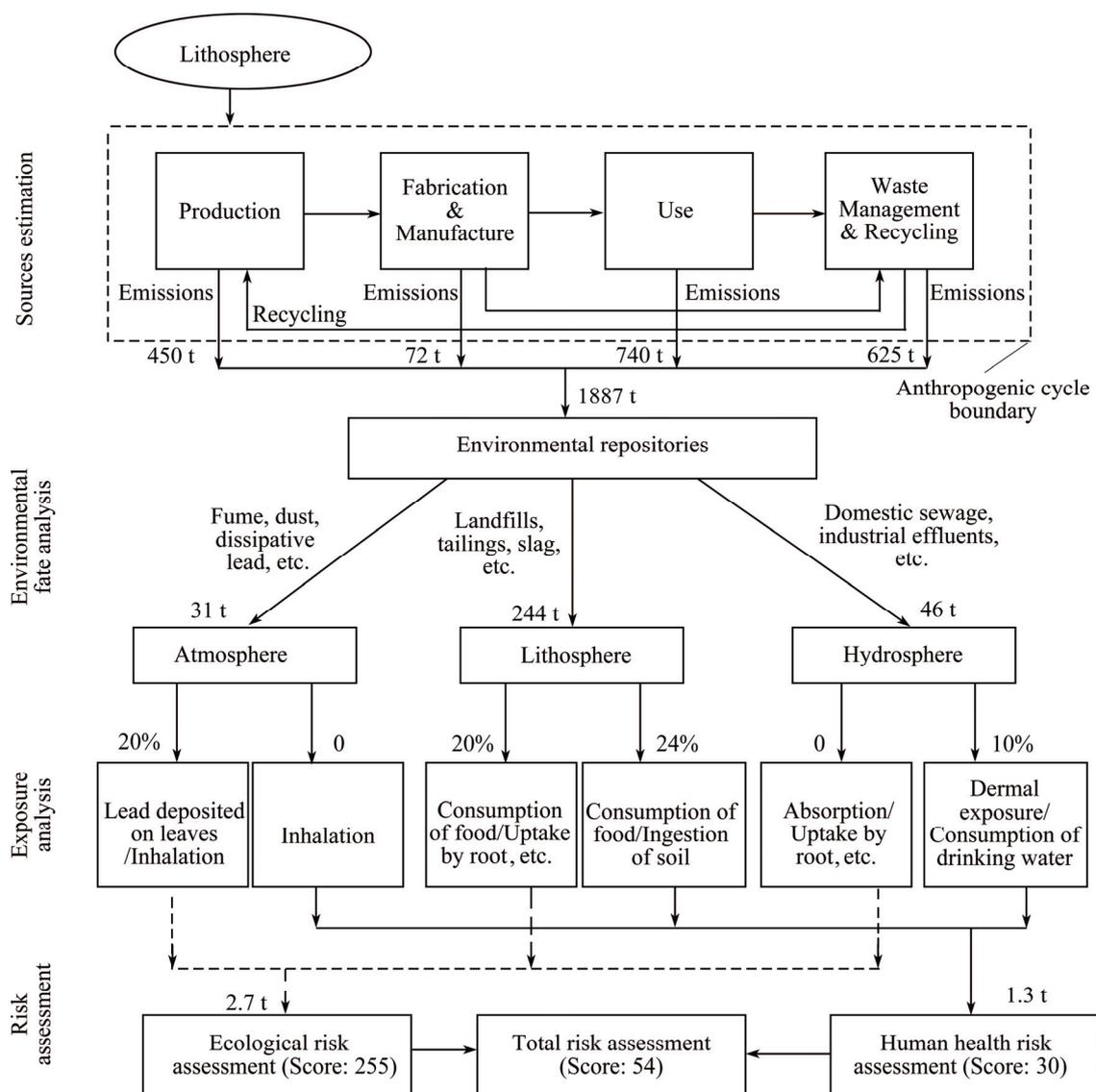


Fig. 4 Risk assessment of lead emissions in China in 2010

4) Uncertainty in factor estimation. Because this work tries to estimate the lead risk with a new method, the studies that can be referred to are limited. The determination of factors such as the decrease factors and exposure factors are attempted, and their precision remains to be checked in the future.

5) Uncertainty in the unit risk scores. The scores in the IRCH ranking system does not cover all the chemicals, and the estimation for the unlisted ones cannot be as precise as the listed ones.

4 Conclusions

1) A model for lead risk assessment is established, and the human health risk, ecological risk and total risk are assessed. The source emissions were 1887 t in 2010 for China. The lead that reached human body was quantified to be 1.3 t (0.9 mg/ca) while the lead that reached ecosystem was 2.7 t. Based on the unit risk score obtained from the IRCH ranking system, the human health risk is calculated to be 30, the ecological risk is 255 and the total risk is 54.

2) In terms of life cycle stages, lead emissions mainly come from the Use stage, while lead emissions reached human and ecosystem mainly come from the Waste Management & Recycling and Production stages. These two stages also contribute most to the total risk.

3) In terms of chemical forms, the most chemicals discharged from the source are PbO and PbSO₄. The most significant chemical for the health risk is PbO while the major chemical for the ecological risk is PbSO₄. For the total risk assessment, PbSO₄, PbO and Pb altogether contribute 71%, which implies that those three forms should be taken priority for lead pollution control.

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铅元素人为循环释放物的风险评价

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摘 要: 开展从源头释放到风险产生全过程的评价可有效指导铅污染的控制。通过建立包含释放物源头、环境过程、暴露途径及风险值计算的铅风险评价的模型, 对人为循环中铅源头释放物的健康风险、生态风险及综合风险进行评价。采用系数估算值来表征铅释放物在环境中衰减和暴露水平。结果表明: 2010 年中国所产生的 1887 t 铅释放物(基于前期研究结果), 经过环境衰减和暴露过程后, 可形成健康风险的铅释放物量为 1.3 t(人均水平为 0.9 mg/ca), 而可形成生态风险的铅释放物量为 2.7 t。铅释放的源头集中在使用阶段, 而产生的风险主要集中在废物管理与回收阶段和生产阶段。从形态来看, 产生健康风险最多的形态为 PbO, 而 PbSO₄ 为造成生态风险最多的化合物。铅释放物中的 PbSO₄、PbO 和 Pb 形态所导致的风险占总风险的 71%, 在风险管理中应关注这三种形态。

关键词: 铅; 源头; 健康风险; 生态风险; 综合风险; 生命周期

(Edited by Mu-lan QIN)