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Lead anthropogenic transfer and transformation in China

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Abstract: Information on lead redistribution and speciation changes in anthrosphere can help to analyze the whole lead cycle on the earth. Lead life cycle was traced based on the concepts of anthropogenic transfer and transformation. Lead transfer and the distribution of chemical species throughout the anthropogenic flow were identified in 2010 in China. The results show that 1.85 Mt lead ore was consumed (besides 1.287 Mt imported concentrated ore and 1.39 Mt lead scraps. After undergoing transformations, 3.53 Mt lead entered end services in chemical species of Pb, PbO_2 and $PbSO_4$, altogether accounting for over 80% of the total lead products. Finally, 2.10 Mt ore was emitted into the environment in such species as $PbSO_4$ (26%), PbO (19%) and Pb (15%). Lead transfer begins in primary raw material sectors, and then transfers to manufacturing sectors. Lead provides services mainly in such industrial sectors as transportation, electrical power and buildings or construction.

Key words: lead element; transfer; transformation; anthrosphere; redistribution; industrial sectors; chemical species; life cycle

1 Introduction

Humans have increasingly altered the physical and chemical processes on the earth's surface over the last several centuries. However, it is only within the past several decades that the impact of humans on the earth has begun to widely attract the attention of scientists [1]. Human activities have changed the mobilization and transformation processes of many elements in the nature, including the heavy metals such as lead [2]. Specifically, the scale of anthropogenic lead cycle is much greater than the natural cycle [1]. As anthropogenic activities have increasingly disturbed the earth's natural system, lead anthropogenic transfer and transformation become integral parts of its biogeochemical cycle. As one of the most poisonous metals in human civilization, lead poses great threats to eco-security as well as human health [3]. Therefore, research on lead transfer and transformation is desperately pressing. Currently, many studies have outlined the characteristics of anthropogenic lead cycle by quantifying the flows of lead [4−7]. Those reports have established a foundation for a further study of lead cycle whereas some shortcomings still exist. Studies on transfer and transformation seem to be limited at present. Some studies on lead transfer have mainly focused on the transfer processes occurring in nature, such as the metal transfer in soil-plant system and the mass transfer between different media [8−10], and others focus on toxicology studies [11,12]. Lead transformation studies have been generally limited to the changes caused by technical innovations [13,14] or natural process such as that in the soil [15,16]. Therefore, lead redistribution and speciation after anthropogenic input are not fully understood, and this knowledge is essential to clarify human interference as well as understand the complex processes of pollution formation.

We pay special attention to the anthropogenic lead flow in China in 2010. China has become one of the largest lead producer and consumer all over the world [17]. Additionally, with the booming economy and rising social demands, the scale of lead transfer and transformation in China will continue to increase in future [18]. Therefore, lead cycle in China is a representative case study to examine lead redistribution and speciation change after anthropogenic input.

2 Methodology

2.1 Basic concepts

Practically, the changes such as product features, chemical species or locations [19] occur consistently in

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the anthropogenic lead flow. We will explain some key concepts and give a brief retrospect in the following part because those concepts are not widely understood now.

2.1.1 Anthropogenic transfer

Anthropogenic transfer normally refers to the mobilization and corresponding virtual redistribution of substances in the anthrosphere. They are driven by the social demands and are accomplished under human activities [20]. The natural transfer of pollutants refers to the moving processes of pollutant in the environment, which normally leads to the enrichment, dispersion or disappearance of the pollutants [21]. The differences between the anthropogenic transfer and the natural transfer are as follows: 1) the driving forces behind anthropogenic transfer are social demands other than the natural forces, which motivate the natural transfer; 2) the anthropogenic transfer occurs in the hybrid socio-economic system, although this system is often related to the natural environment.

Anthropogenic transfer can be classified into two categories: transfers in physical space and in virtual space. Physical transfer involves the transfers between different regions or the earth's spheres, i.e., the lithosphere, hydrosphere, atmosphere and anthrosphere. For example, lead is transferred from the lithosphere to the anthrosphere after lead ores are mined from the earth. The virtual space transfer refers to the transfer between different industrial sectors, which are conceptional but essential for knowing lead movement in the whole society. In this work, we will focus on the virtual anthropogenic transfer, regardless of the plentitude of studies on physical transfer [22,23].

In this work, we will focus on the transfer in the virtual space. Based on lead life cycle, lead transfer can be further classified into the transfer at production, fabrication and manufacture (F&M), use and waste management and recycling (WMR) stages. Each transfer at a certain life stage can be further subdivided into different industrial sectors. Also, according to different purposes, lead transfer can be divided into the flow to satisfy production needs, the flow to manufacture product and the flow to meet the demand of humans as a finished product. For example, lead flow from the lithosphere to refining industry belongs to the flow to satisfy production needs.

In the framework (Fig. 1), the detailed acronyms for

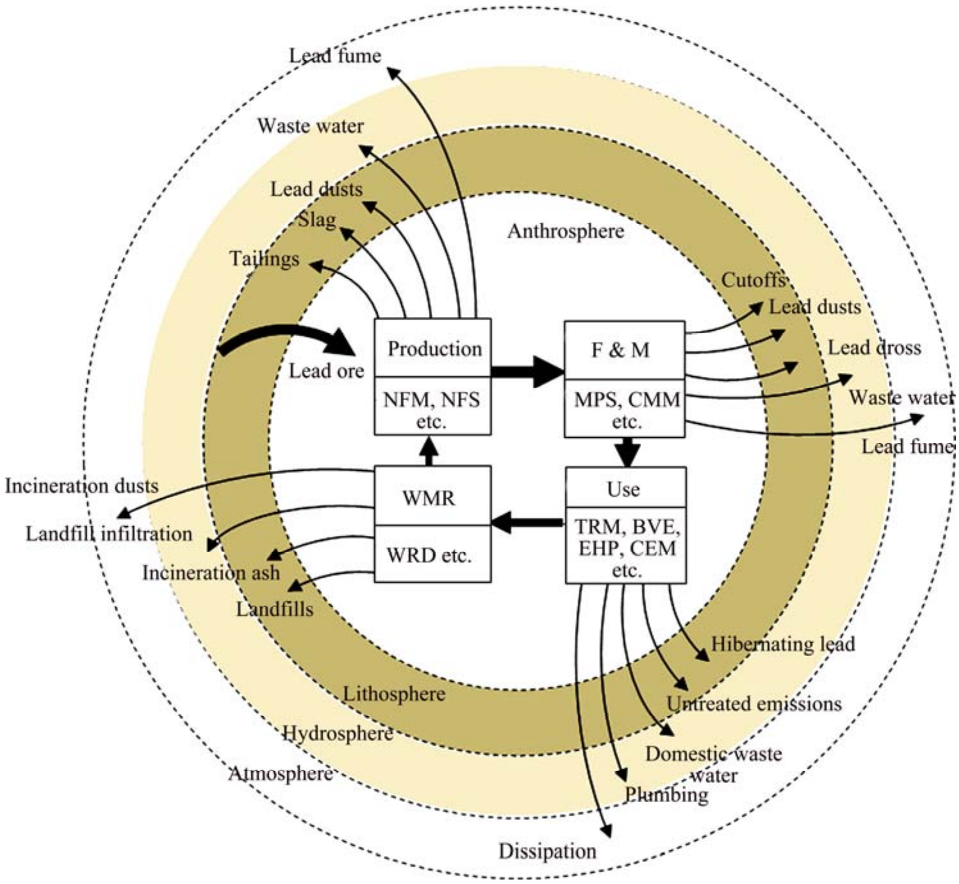

Fig. 1 Schematic diagram of anthropogenic transfer of lead (Acronyms for industrial sectors: NFM—Nonferrous metal mining; NFS—Nonferrous metal smelting; MPs—Metallic products and parts supplement; CMM—Chemical material manufacturing; TRM—Transportation manufacturing; BVE—Building and vihicle engineering; EHP—Electricity and heating power; CEM—Computer and electronics manufacuturing; WRD—Waste resources and discard recycling)

the industrial sectors are cited based on Ref. [24]. Lead goes through life stages while transferring in various industrial sectors. Moreover, lead losses are emitted into the earth's spheres. For example, tailings and slag are discharged from production. Lead dust and waste water are released into soil, atmosphere or water from F&M. Hibernating lead refers to lead that is not finally disposed of, but no longer serving any useful function [5].

2.1.2 Anthropogenic transformation

Anthropogenic transformation refers to the process where chemical species of a material are changed into another or change into several other species, and the new species are meant to satisfy specific human demands. Practically, these processes can be achieved by various engineering techniques [20]. The natural transformation of pollutants refers to the physical, biological or chemical reactions where a substance changes into another [21]. The differences can be figured out as follows: 1) the natural one is affected by many factors, such as the temperature, pH, soil or water properties and other anions [15], while anthropogenic flow is mainly affected by social factors; 2) the natural one tends to form stable substances while anthropogenic transformation aims at serving human beings.

Generally, chemical species refer to the chemical species, but they can also refer to the mineralogical, molecular, or ionic type [25]. In this work, we analyze the chemical species although they are combined with other materials. For example, PbO can exist in acidic battery paste but is also widely used as a cosolvent in alloys.

2.2 Analysis of lead anthropogenic transfer and transformation

Overall, lead life cycle is traced with material flow analysis to understand the transfer and transformation. Then, lead transfer between different industrial sectors is studied. Finally, possible transformation is identified with a physicochemical analysis to find the possible changes in lead species during the life cycle.

2.2.1 Tracing lead life cycle

The boundary in this work is the system made of all the four lead life cycle stages. The geographical boundary is the mainland China and the reference year is 2010. Lead life cycle starts with the mining of lead resources, continues with the lead fabrication and manufacturing into products or semi-products, which enter the use stage serving humans, and ends up with lead deposited in landfill after recycling. In short, it contains all the stages that metal experiences from the state of resource to waste. Previous research on lead paid close attention to the quantities of the multilevel lead cycle such as lead in-use stock and lead losses to the environment [26−28], which can be useful in tracing lead. By material flow analysis, which is widely applied to characterizing the flow in a well-defined system [29], we tried to find out the mass of each flow in lead cycle based on data from the database or estimation. The flows needed to be determined can be seen from the schematic diagram, which were established in STAF project in 2006 as Fig. 1 in Ref. [5]. Additionally, mass conservation law must be observed. Tracing lead life cycle helps to determine the exact stage where lead transfer occurs. Although the import and export cannot be excluded from the flow (they are important for the balance of the flows), we mainly focus on the domestic ones in China.

2.2.2 Analysis of anthropogenic transfer

Lead transfers in different industrial sectors while flowing through the anthropogenic cycle. Principal lead products are lead acid batteries, pigments, alloys, ammunition, cable sheathing and additives. We identified where lead products fulfill their function based on the necessary information and the general knowledge. We followed lead transfer into different sectors and traced lead redistribution in the human society. In this work, we chose the industrial sectors according to the China's National Standard of Industial Classification, GB/T 4754−2011 [30].

2.2.3 Analysis of anthropogenic transformation

Lead products cannot fulfill their functions without certain chemical species. It is already known that anthropogenic input acts as the dominant driving force behind the transformation. To analyze species transformation, we firstly identified the chemical species before they underwent the industrial processes, and then referred to handbooks or technical references to find out the probable chemical reactions that happened in the industrial processes, which helped us to predict the possible outcomes of reactions. Furthermore, during those chemical reactions, we also tried to analyze the emissions from anthrosphere. To conclude, we focused on the initial species of lead compounds as well as the endpoints of the industrial processes while analyzing the possible industrial emissions during those reactions.

2.3 Data sources

Some basic data are essential to determine lead flows, which are already available. Their values and sources are shown in Table 1. Those data are the basis for the further calculation and estimations.

3 Determination of lead flows and species

3.1 Lead flow quantities in anthropogenic cycle

Although some lead flows are accessible, some others cannot be acquired precisely for the moment such as the lead flow in China in 2010. In this work, the

Table 1 Basic data for calculation of anthropogenic lead cycle and their data sources

Parameter	Value	Data source
Mass flow of refined lead production/kt	4200	[31]
Mass flow of primary refined lead/kt	2840	[31]
Mass flow of secondary refined lead/kt	1360	[31]
Mass flow of domestic lead ore/kt	1850	[31]
Mass flow of lead consumption/kt	3950	[18]
Mining efficiency	0.84	[18]
Mass flow of primary refining efficiency	0.96	[18]
Mass flow of lead in net import of lead ore/kt	1287	[18]
Mass flow of lead in net export of products/kt	50	[18]
Mass flow of lead in net import of scrap/kt	0	[18]

quantities of those flows were determined.

The recycling flows are important in anthropogenic lead cycle. In secondary production, the scraps entering secondary production are estimated as the secondary refined lead (1360 kt) divided by the recovery rate (98% [5]). F&M scrap occupies approximately 4% of the total scrap [32]. Apart from the recycled flows, lead products are manufactured with the fabrication efficiency of 91% [5], and some lead products become in-use stock, for which the accumulation rate is 0.18 [6]. The net import of lead ore and net export of lead products are 1287 kt and 50 kt, respectively, while the net export or import of lead scraps is zero because the import and export of lead scraps are strongly restricted according to the Law of the People’s Republic of China on the Prevention and Control of Environmental Pollution by Solid Waste and Basel Convention on the Control of Transboundary Movements of Hazardous Waste and Their Disposal. In addition, lead sludge and lead slag are listed among the Catalogue of Commodities Forbidden to Import (the third batch and the fourth batch). And also, the scarp import and export data are not included in the China Nonferrous Metals Industry Yearbook [18]. Although the life spans of lead products may affect the quantities at the use stage [26], in this work, we mainly focused on a static analysis of lead transfer and transformation in a specific year, and a dynamic analysis of lead in-use stock is not included.

Lead losses from the anthrosphere differ greatly in every stage of the life cycle. Tailings are estimated from the lead ores multiplied by 1 minus the mining rate, and refining slag equals the primary or secondary refined lead multiplied by 1 minus the primary or secondary refining rate. Lead losses from F&M are less than 5% of the total losses [6]. The high proportion of dissipative lead is responsible for the heavy loss in use, which is estimated to be about 30% of the total losses [32]. For every kilogram of lead consumed, 0.5 kg is assumed to be lost to the environment [3]. Following previous study [3,32], the total losses to environment are estimated to be 2.1 Mt. Finally, with mass conservation law, we can work out all the data, such as the lead entering WMR and the losses to the environment from WMR. The lead flow in 2010 in China is shown in Fig. 2.

3.2 Lead transformation at production stage

At the production stage, for lead ore, galena (PbS), cerusite ($PbCO_3$) and anglesite ($PbSO_4$) are in the raw materials [33], and galena accounts for 75% (mass fraction), cerusite constitutes 15% (mass fraction), and the left are anglesite or other species [34].

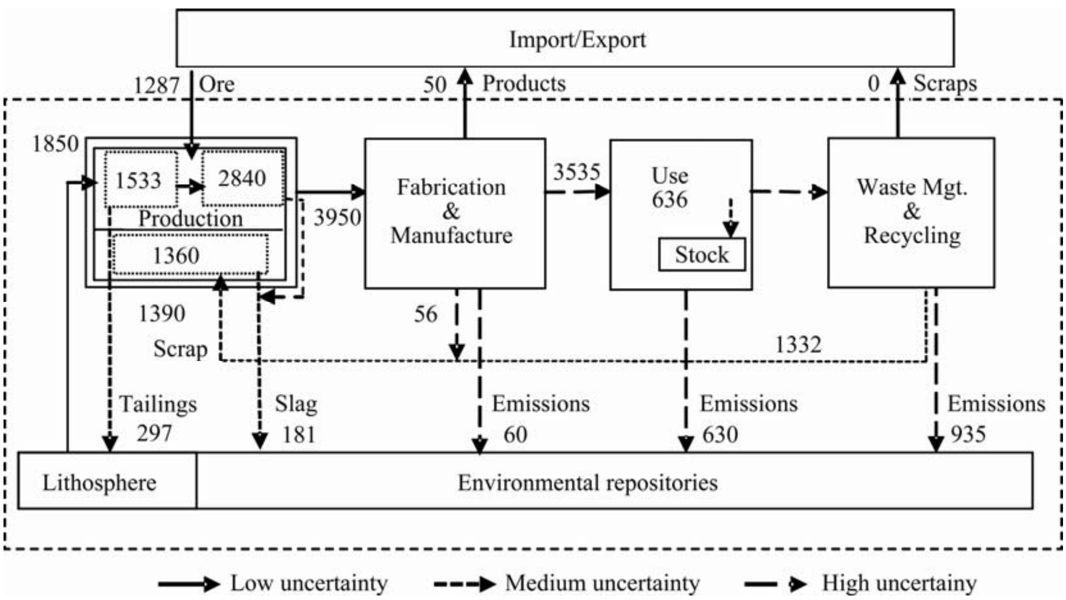

Fig. 2 Anthropogenic flow of lead in 2010 in China (unit: kt)

Wastes are discharged during the production process, including primary refining, secondary refining and mining activities. In production, mining contributes a lot to the emissions into the environment [35]. The mass proportion of those wastes can be referred to from the cleaner production audit report [36], on-site monitoring or the lead mass balanced sheet [28]. The primary species in wastes can be identified, with $PbSO_4$ being the main species in tailing, PbO and PbS in primary lead refining process and PbS and PbO_2 in secondary lead refining. We can get the exact amount of the species by multiplying the total quantities of tailings, primary lead slag and secondary lead slag by the ratio of the species in the wastes [37], and then sum up the same species.

3.3 Lead transformation at F&M stage

At the F&M stage, the principal lead products and the species in them are focused on to get the species distribution. Lead acid batteries, lead oxidants, lead alloys, and rolled and extruded products altogether account for 94% of the total, of which batteries constitute 73%, and lead alloys occupy 8% [38]. As for the chemical species, in lead acid batteries, the active material in the positive pole is mainly PbO_2 while the negative pole consists of spongy lead; in lead oxidants, the species are mainly PbO, Pb_3O_4 and Pb_2O_3; lead alloys, and rolled and extruded products are mainly metallic lead. With the proportion of species, an approximate speciation can be obtained at F&M stage in the anthrosphere.

Lead losses are discharged during the manufacturing. For lead acid batteries, the emissions are largely PbO, Pb and $PbSO_4$ [39]. For lead oxidants production, lead oxidants are usually produced with the metal oxidation method and PbO, Pb, Pb_2O_3 and Pb_3O_4 are lost. For lead alloy smelting, PbO and Pb are emitted in dross or fumes [40].

3.4 Lead transformation at use and WMR stage

A similar calculation was carried out for speciation at use and WMR stages. The species at those two stages may change and differ from those at production and F&M stage. In lead acid batteries, $PbSO_4$ is formed when the battery is discharged. A large part of lead oxidants consumed is PbO (massicottite) and Pb_3O_4 (red lead). Lead pigments and additives are mainly lead salt compounds, such as $PbCO_3$ and $PbCl_2$. The lead species at WMR stage show the species of discarded products whereas the losses from WMR show the species in landfill.

Use losses to the environment are dissipative in nature. Batteries discarded without treatment are considered to be the losses during use. The proportions of Pb, PbO_2 and $PbSO_4$ are 30%, 30% and 40%, respectively. For lead oxidants, losses in PbO, Pb_2O_3 and Pb_3O_4 are 60%, 30% and 10%, respectively. For lead additives, $PbCO_3$ and $PbCl_2$ constitute about 30% of all emissions, with the left being other species [37]. In waste batteries, $PbSO_4$, PbO_2, Pb and PbO account for approximately 50%, 20%, 17% and 13% of the total lead in batteries, respectively [41]. Lead oxidants will release complex compounds when they are treated: Pb_3O_4 is released as the ingredient of red lead paint; $PbCO_3$ in lead subcarbonate is also emitted; chemicals such as Pb_2O_3 are released from incineration. In China, with the high content of chlorine and sulfur in wastes, during incineration lead residues and fly ash are usually discharged with $PbCl_2$, $PbSO_4$ and lead oxidants, and the mass ratio of chloride to sulfide is 2:1 [42].

4 Results and discussion

4.1 Anthropogenic transfer in anthropogenic lead cycle

Lead anthropogenic transfers play an important role in the social-economic interaction. Lead transfers in a multitude of industrial sectors. In production, lead is transported from the industrial sector of NFM to NFS. At F&M stage, refined lead is manufactured into products after being transmitted to the industrial sector of MPM or CMM (the acronyms can be referred to in Fig. 1). During use, lead products, including lead batteries, lead alloys and building materials, realize their functions after entering into the industrial sectors, such as TRM, BVE and EHP. Finally, discarded products are recycled or disposed of in WRD.

In conclusion, lead firstly moves into primary industrial sectors of raw material processing, such as lead mining and refining sectors, and then enters secondary manufacturing sectors. Then, lead products provide services mainly in such industrial sectors as transportation, electrical power and buildings and construction. Finally, lead ends its transfer process when discarded lead is recycled or put into landfilling.

Lead transfer is reflected directly by the change of flux. The refined lead production was 4.20 Mt in 2010 [18], with 1.85 Mt lead ore being mined domestically. Then, 3.95 Mt was consumed, with 3.53 Mt entering end services and approximately 2.10 Mt lost into environmental repositories (Fig. 3), namely the atom- sphere, hydrosphere and pedosphere. WMR emissions are higher in mass than other emission flows. Based on all the data of lead flow, it can be concluded that in the anthropogenic cycle for every 1 kg of lead consumed, 0.89 kg lead comes into the domestic market, 0.47 kg lead needs to be mined from the lithosphere and 0.53 kg lead is lost to the environment.

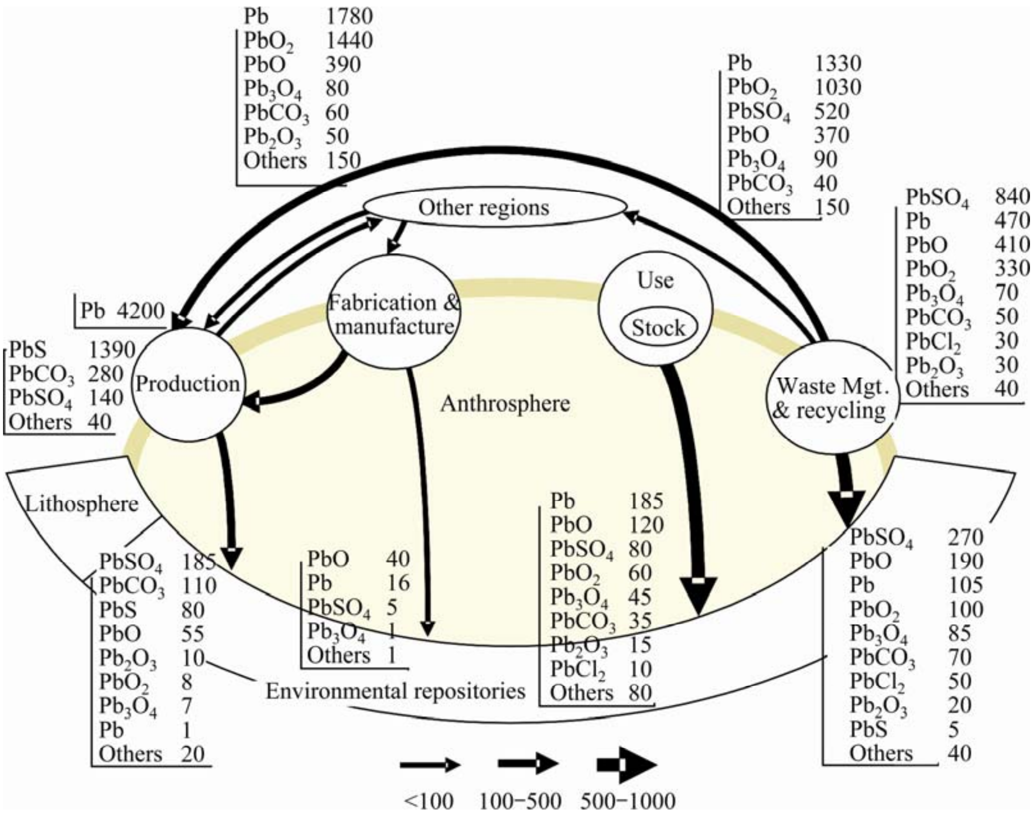

Fig. 3 Schematic diagram of lead anthropogenic tranfer and transformation of China in 2010 (unit: kt/year)

4.2 Implications on resources and environment

Overall lead in the anthrosphere transfers in the following direction: resources — end services — environmental repositories. Lead is mined from natural resources, fabricated into lead products, which enters end services for human welfare, and finally discarded into the environment after being used. It can be seen from Table 2 that there is a huge difference among the lead species in resources, end services and environmental losses. PbS, $PbCO_3$ and $PbSO_4$ are the primary chemical species in resources, in which PbS accounts for as high as 75%, with $PbCO_3$ and $PbSO_4$ occupying 15% and 8%, respectively. After being transformed into various products, lead serves human society largely in the chemical species of Pb (38%), PbO_2 (26%) and $PbSO_4$ (15%). Lead losses are released from the anthropogenic cycle as $PbSO_4$ (26%), PbO (19%), Pb (15%) and $PbCO_3$ (10%), among which $PbSO_4$ probably comes from tailings and waste lead acid batteries, and PbO is from lead chemicals and batteries. The toxicity of those species varies a great deal and one of the most suitable hazard score ranking was given by the Indiana Relative Chemical Hazard (IRCH) Ranking System [43].

Rules of lead transfer and transformation could identify the adverse influences on the resources or environment and allow a better control of lead inflows and outflows. Overdoses may compromise the future availability for human utilization and improper disposal could result in a huge health risk in potential, such as the negative effect on developmental intelligence quotient (IQ) and blood lead levels [44]. Therefore, knowledge of lead life cycle could help to improve or even provide guidance for human activity so as to alleviate the negative effects on lead flow. For example, efficiencies, such as the mining efficiency or fabrication efficiency, should be improved to ensure that less is lost to the environment and more enters the products. Human should also be sparing when exploiting the resources, because more mining may lead to more environmental losses [32]. At last, the mass of lead flow can be less if the demand for lead is reduced. Therefore, human should actively seek for lead substitution to reduce the amount of lead consumed and thus reduce the pollution or help resource conservation.

4.3 Data uncertainty

Due to uncertainty, the lead cycle is not entirely correct and complete. There is an inconsistency in data source with varying reliability-data for the refined lead production, primary or secondary lead and mined ore are from the USGS, while the others are from the CNMIY (China Nonferrous Metals Industry Yearbook). Some flow mass may be not accurate enough, especially the emission flows, as they are based on previous research

Table 2 Distribution of lead chemical species in lead resources, end services and losses

Item	Chemical species	Mass flow/kt	Percent of total/%
Resource	PbS	1390	75
	$PbCO_3$	280	15
	$PbSO_4$	150	8
	Others	30	2
	Total	1850	100
End service	Pb	1330	38
	PbO_2	1030	29
	$PbSO_4$	520	15
	PbO	370	10
	Pb_3O_4	90	3
	$PbCO_3$	45	1
	Others	150	4
	Total	3535	100
Environmental loss	$PbSO_4$	540	26
	PbO	405	19
	Pb	307	15
	$PbCO_3$	215	10
	PbO_2	168	8
	Pb_3O_4	138	7
	PbS	85	4
	Pb_2O_3	45	2
	$PbCl_2$	60	3
	Others	141	7
	Total	2104	100

work. And some empirical information is used, such as fabrication efficiency, the mass ratio of F&M scrap to total scrap.

Although the best estimations are made to study lead anthropogenic transformation, the species analysis is far from accurate. The absent information was estimated, such as the species percentage of lead oxidants, lead pigments and additives. Therefore, high uncertainty exists except for lead acid batteries, which have available information like the species in F&M losses, and in new or waste battery.

5 Conclusions

1) Lead anthropogenic transfer and transformation greatly differ from the traditional one in geochemistry as it focuses on the relationship between anthropogenic input and material redistribution or species change.

2) Lead transfers in different industrial sectors. Lead products provide services mainly in such industrial sectors as transportation, electrical power and construction. Also, lead transfer can be shown by the change of flux. For every 1 kg of lead consumed, 0.89 kg comes into the domestic market, 0.47 kg lead needs to be mined from the lithosphere and 0.53 kg lead is lost to the environment.

3) Lead anthropogenic transformation refers to the chemical species change in the anthropogenic cycle. For China, lead ores are primarily in the specie of PbS (75%). Lead products in end services are largely in chemical species of Pb, PbO_2 and $PbSO_4$ (over 80% of the total). Also, lead losses emitted into the environment are primarily in such species as $PbSO_4$ (26%), PbO (19%) and Pb (15%).

References

[1] SEN I S, BERNHARD P E. Anthropogenic disturbance of element cycles at the earth's surface [J]. Environmental Science and Technology, 2012, 46(16): 8601−8609.

[2] RAUCH J N, PACYNA J M. Earth's global Ag, Al, Cr, Cu, Fe, Ni, Pb and Zn cycles [J]. Global Biogeochemical Cycles, 2009, 23(2): 1−16.

[3] SMITH D R, FLEGAL A R. Lead in the biosphere: Recent trends [J]. Ambio, 1995, 24(1): 21−23.

[4] ELSHKAKI A, van der VOET E, HOLDERBEKE M V, TIMMERMANS V. Long-term consequences of non-intentional flows of substances: Modeling non-intentional flows of lead in the Dutch economic system and evaluating their environmental consequences [J]. Waste Managament, 2009, 29(6): 1916−1928.

[5] MAO J S, DONG J, GRAEDEL T E. The multilevel cycle of anthropogenic lead. I: Methodology [J]. Resources Conservation and Recycling, 2008, 52(8−9): 1058−1064.

[6] MAO J S, DONG J, GRAEDEL T E. The multilevel cycle of anthropogenic lead. II: Results and discussion [J]. Resources Conservation and Recycling, 2008, 52(8−9): 1050−1057.

[7] MAO J S, CAO J, GRAEDEL T E. Losses to the environment from the multilevel cycle of anthropogenic lead [J]. Environmental Pollution, 2009, 157(10): 2670−2677.

[8] BI X Y, REN L M, GONG M, HE Y S, WANG L, MA Z D. Transfer of cadmium and lead from soil to mangoes in an uncon taminated area, Hainan Island, China [J]. Geoderma, 2010, 155(1−2): 115−120.

[9] GUILLÉN J, BAEZA A, ONTALBA M A, MÍGUEZ M P. ^{210}Pb and stable lead content in fungi: Its transfer from soil [J]. Science of the Total Environment, 2009, 407(14): 4320−4326.

[10] KONDO A, YAMAMOTO M, INOUE Y, ARIYADASA B H. Evaluation of lead concentration by one-box type multimedia model in Lake Biwa-Yodo River Basin of Japan[J]. Chemosphere, 2013, 92(5): 497−503.

[11] HALLÉN I P, JORHEM L, OSKARSSON A. Placental and lactational transfer of lead in rats: A study on the lactational process and effects on offspring [J]. Archives of Toxicology, 1995, 69(9): 596−602.

[12] LIN C M, DOYLE P, WANG D L, HWANG Y H, CHEN P C. The role of essential metals in the placental transfer of lead from mother to child [J]. Reproductive Toxicology, 2010, 29(4): 443−446.

[13] VOLPE M, OLIVERI D, FERRARA G, SALVAGGIO M, PIAZZA S, ITALIANO S, SUNSERI C. Metallic lead recovery from lead−acid battery paste by urea acetate dissolution and cementation on iron [J]. Hydrometallurgy, 2009, 96(1−2): 123−131.

[14] ZHU X F, YANG J K, GAO L X, LIU J W, YANG D N, SUN X J, ZHANG W, WANG Q, LI L, HE D S, KUMAR R V. Preparation of lead carbonate from spent lead paste via chemical conversion[J]. Hydrometallurgy, 2013, 134−135: 47−53.

[15] BIRKEFELD A, SCHULIN R, NOWACK B. In situ transformations of fine lead oxide particles in different soils [J]. Environmental Pollution, 2007, 145(2): 554−561.

[16] CAO X D, MA L Q, CHEN M, HARDISON D W, HARRIS W G. Lead transformation and distribution in the soils of shooting ranges in Florida, USA [J]. Science of the Total Environment, 2003, 307(1−3): 179−189.

[17] ZHANG X W, YANG L S, LI Y H, LI H R, WANG W Y, YE B X. Impacts of lead/zinc mining and smelting on the environment and human health in China [J]. Environmental Monitoring and Assessment, 2012, 184(4): 2261−2273.

[18] Editorial Staff of the China Nonferrous Metals Industry Yearbook. China nonferrous metals industry yearbook, 2000−2012 [M]. Beijing: China Nonferrous Metals Industry Yearbook Press. (in Chinese)

[19] MAO Jian-su, MA Lan, LIANG Jing. Changes in the functions, species and locations of lead during its anthropogenic flows to provide services [J]. Transactions of Nonferrous Metals Society of China, 2014, 24(1): 233−242.

[20] MAO J S, MA L, NIU J P. Anthropogenic transfer & transformation of heavy metals in anthrosphere: Concepts, connotations and contents [J]. International Journal of Earth Sciences and Engineering, 2012, 5(5): 1129−1137.

[21] DAI Shu-gui. Environmental chemistry [M]. Beijing: Higher Education Press, 2006: 14. (in Chinese)

[22] NIISOE T, NAKAMURA E, HARADA K, ISHIKAWA H, HITOMI T, WATANABE T, WANG Z, KOIZUMI A. A global transport model of lead in the atmosphere[J]. Atmospheric Environment, 2010, 44(14): 1806−1814.

[23] XU H, SUN Y Y, DING Z H, ZHANG Y, WU J C, LIAN H Z, WANG T J. Lead contamination and transfer in urban environmental compartments analyzed by lead levels and isotopic compositions [J]. Environmental Pollution, 2014, 187: 42−48.

[24] MAO J S, DU Y C, XU L Y, ZENG Y. Quantification of energy related industrial eco-efficiency of China [J]. Frontiers of Environmental Science and Engineering in China, 2011, 5(4): 585−596.

[25] ZHENG G D, FU B H, TAKAHASHI Y, KUNO A, MATSUO M, ZHANG J D. Chemical speciation of redox sensitive elements during hydrocarbon leaching in the Junggar basin, northwest China[J]. Journal of Asian Earth Sciences, 2010, 39(6): 713−723.

[26] MAO J S, GRAEDEL T E. Lead in-use stock [J]. Journal of Industrial Ecology, 2009, 13(1): 112−126.

[27] GERST M D, GRAEDEL T E. In-use stocks of metals: Status and implications [J]. Environmental Science and Technology, 2008, 42(19): 7038−7045.

[28] ELSHKAKI A, van der VOET E, van HOLDERBEKE M, TIMMERMANS V. The environmental and economic consequences of the developments of lead stocks in the dutch economic system[J]. Resources Conservation and Recycling, 2004, 42(2): 133−154.

[29] GLÖSER S, SOULIER M, TERCERO ESPINOZA L A. Dynamic analysis of global copper flows: Global stocks, postconsumer material flows, recycling indicators, and uncertainty evaluation [J]. Environmental Science and Technology, 2013, 47(12): 6564−6572.

[30] Statistic Design Management Department of National Bureau of Statistics of PRC. GB/T 4754−2011 for national economic activities industrial classification [S]. (in Chinese)

[31] USGS (U.S. Geological Survey). Minerals yearbook — Lead [EB/OD]. 2013−10−09. http://minerals.usgs.gov/minerals/pubs/commodity/lead/.

[32] LIANG Jing, MAO Jian-su. A dynamic analysis of environmental losses from anthropogenic lead flow and their accumulation in China [J]. Transactions of Nonferrous Metals Society of China, 2014, 24(4): 1125−1133.

[33] WANG Ji-kun. Handbook of lead and zinc smelting production technology [M]. Beijing: Metallurgical Industry Press, 2012: 30. (in Chinese)

[34] YU Xue, CHEN Hong. The experimental study of flotation on improving recovery of a lead ores [J]. Non-Ferrous Mining and Metallurgy, 2005, 21(6): 18−20. (in Chinese)

[35] ZHANG Xiu-wu, YANG Lin-sheng, LI Yong-hua, LI Hai-rong, WANG Wu-yi, GE Quan-sheng. Estimation of lead and zinc emissions from mineral exploitation based on characteristics of lead/zinc deposits in China [J]. Transactions of Nonferrous Metals Society of China, 2011, 21(11): 2513−2519.

[36] ZHAO Xiao-sheng. Cleaner production evaluation of innovations on ISP lead and zinc smelting [J]. Engineering Design and Research, 2007, 123: 11−19. (in Chinese)

[37] LIANG Jing, MAO Jian-su. Speciation analysis of lead losses from anthropogenic flow in China [J]. Environmental Sciences, 2014, 35(3): 1191−1197. (in Chinese)

[38] WANG Ye. Development and perspectives of Chinese lead industry [J]. Nonferrous Metals Engineering, 2011(1): 27−29. (in Chinese)

[39] LIU Guang-lin. Introduction of the technology on lead acid battery [M]. Beijing: China Machine Press, 2011. (in Chinese)

[40] Editorial Board of Non-ferrous Metals Industry Analysis. Series of non-ferrous metals industrial analysis [M]//Chemical phase analysis for ore and industrial products. Beijing: Metallurgical Industry Press, 1992: 255−258. (in Chinese)

[41] ZHOU Zheng-hua. Pollution-free pyrometallurgical regeneration of lead and its alloys from scrapped batteries [J]. Shanghai Nonferrous Metals, 2002, 23(4): 157−163. (in Chinese)

[42] SHI D Z, WU W X, LU S Y, CHEN T, HUANG H L, CHEN Y X, YAN J H. Effect of MSW source-classified collection on the emission of PCDDs/Fs and heavy metals from incineration in China [J]. Journal of Hazardous Material, 2008, 153(1−2): 685−694.

[43] ECKELMAN M J, GRAEDEL T E. Silver emissions and their environmental impacts: A multilevel assessment [J]. Environmental Science and Technology, 2007, 41(17): 6283−6289.

[44] CARRINGTON C D, BOLGER P M. Toxic metals: Lead [J]. Encyclopedia of Food Safety, 2014, 2: 349−351.

中国铅元素的人为迁移与转变

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摘 要：研究铅的人为迁移所导致的再分布以及形态转变有助于补充和完善铅的整个地球化学循环过程。在回顾铅人为迁移和转变概念基础上，通过追踪铅元素的产品生命周期过程，研究中国 2010 年的铅元素人为循环中的人为迁移及各化学形态的转变。结果表明，2010 年消耗国内 1.85 Mt 铅矿石，外加 1.287 Mt 进口铅精矿和 1.39 Mt 废铅，经过人为转变后共有 3.53 Mt 铅进入末端服务，主要以 Pb、PbO_2 和 $PbSO_4$ 等形态(约占 80%)进入产品。并最终向环境中排放 2.10 Mt 释放物，释放物的主要形态为 $PbSO_4$(26%)、PbO(19%)和 Pb(15%)。铅的人为迁移始于原材料部门，然后转入制造部门，而最终服务于交通、电力供应和建筑等工业部门。

关键词：铅元素；迁移；转变；人类活动圈；再分布；工业部门；化学形态；生命周期

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