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Determination of compound wood floor preservative CCA and risk assessment of children's health

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Abstract: *The contents of Cr, As and two other heavy metals Cd and PB in CCA of composite wood flooring were determined by atomic absorption spectrometry and atomic fluorescence spectrometry. Cd, As and Cr in CCA were detected. The concentration of Cd was 0.075~0.5 mg/kg, the maximum concentration of Cr was 2.24~4.09 mg/kg. Health risk assessment of Cd and Cr in laminate flooring was carried out. The non-carcinogenic risk and carcinogenic risk were at low level.*

Keywords: *Atomic absorption; CCA; health risk assessment; natural emission sources*

1. Introduction

CCA is widely used in the preservative treatment of various kinds of wood, which can effectively prevent the harm of decay bacteria, soft rot bacteria, insects, termites and other wood-destroying organisms to wood. The three main active components of CCA (Cu2+, Cr6+, As5+), chromium and arsenic are highly toxic heavy metals, chromium can cause pharyngitis, bronchitis, etc., arsenic mainly damages the skin and causes neuritis. In recent years, considering the potential arsenic exposure hazards and pollution problems of CCA preservatives, since 2004, the United States and European Union countries have gradually restricted the use of CCA preservatives. About 100,000 m^3 of wood treated by CCA in 2008 [1].As CCA treated wood ages, indoor particulate matter contains released arsenic and chromium.

More and more attention has been paid to metal elements in indoor particulate matter. Bai Li [2]'s research results show that the contribution rate of indoor pollution sources of metal elements in indoor PM2.5 is 8.93%.Chen Quan [3] studied that the annual average indoor and outdoor contents of Cr, Cd and As in Zhuzhou exceeded China's Ambient Air Quality Standard (GB3095-2012) and the standard limits of related elements, and believed that the analysis of indoor sources was more complicated. The average human exposure to non-carcinogenic and carcinogenic risks was 8.5 times greater indoors than outdoors.

The method of sampling observation is to use a hollow drill to sample the treated wood and directly observe the color change of the sampled wood section [4-5].

In recent years, people pay more and more attention to the green environmental protection of interior building decoration materials, GB/T31763-2015 copper chromium arsenic (CCA) anticorrosive wood treatment and use specifications, prohibit the use of anticorrosive wood in interior decoration. In this paper, we determined Cr, As and other heavy metals Cd and Pb in the preservatives CCA of composite wood flooring by atomic absorption spectrometry and atomic fluorescence spectrometry for the purpose of detection and risk assessment of preservatives in home decoration materials. The source analysis of heavy metals in indoor air can provide some reference.

2. Materials and methods

2.1 Instruments and reagents

Heavy metal standard products are purchased from the national Standard Material Center, the concentration is 1000ug/ml;

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2.2 Pretreatment of samples

Each sample was accurately weighed 1.00g and put into a 50mL covered beaker, add 20ml nitric acid, place on a 60℃ electric heating plate, keep for 10 minutes, and then raise the temperature to 100℃, keep for about 30 minutes. The color of the digestion solution becomes yellow and transparent, and when the volume is concentrated to about 5ml, add 2ml hydrochloric acid, and remove it when there is no more smoke, cool it to room temperature, and filter it with deionized water to 100ml to be measured.

2.3 Quality assurance and quality control

This experiment strictly followed the QA/QC methods recommended by the US Environmental Protection Agency (USEPA) for sampling, preservation, extraction, and glassware cleaning. Each 10 samples are filled with 1 laboratory blank, and standard substances of known concentrations are added to the samples. The relative recovery rates of the target heavy metals are all between 80-120%.

3. Exposure assessment model

The main exposure route of CCA during infant crawling is percutaneous exposure. With reference to the calculation model of chemical exposure in the United States and the European Union [6-7], the exposure calculation model of percutaneous exposure pathway was established according to the behavioral characteristics of infants and young children.

$$EX_{skin} = \frac{Ci \times S \times G \times ABS}{S \times BW \times AT} \tag{1}$$

Where, in formula (1), EX_skin is the daily percutaneous exposure of human body, mg/(kg•d); Ci is the cumulative exposure concentration of heavy metals during the service life, mg/kg; S is the contact area between wood floor and skin, m^2; G is the quality of wood floor, kg; ABS is the mass fraction of heavy metal transferred to human body through skin. S is the surface area of wood floor, m^2; BW is human body weight, kg; AT is the duration of exposure, d.

The hazard quotient (HQ) represents the non-carcinogenic risk of a population exposed to OPEs through skin and is calculated as shown in equation (2).

$$HQ = EX_{skin}/RfD \tag{2}$$

Where: HQ is the non-carcinogenic risk value of skin exposure to OPEs; RfD is the USEPA reference dose value for each OPEs, ng•(kg•d)−1, HQ<0.1 indicates that the non-carcinogenic risk is small and can be ignored.

$$ILCR = EX_{skin} xCSFO \tag{3}$$

Where: ILCR is the carcinogenic risk value of people exposed to OPEs through skin; The CSFO is a carcinogenic slope factor, mg•(kg•d)−1, used to assess the risk of carcinogenesis associated with exposure to carcinogens or potential carcinogens, as shown in the table 1 below.

Table 1: Carcinogenic risk level

Serial Number	ILCR	cancer risk	Health impact situation
1	$>1\times10^{-6}$	Carcinogenic risk	Affect the normal life of human beings
2	$<1\times10^{-6}$	The cancer risk is negligible	No impact
3	10^{-6}~10^{-4}	A potential cancer risk	There will be diseases
4	$>10^{-4}$	High cancer risk	The health of normal people is affected, and the chronic disease patients are more affected

4. Results and discussion

4.1 The measured content in the sample

In the collected samples, except some samples did not detect Pb, the other three heavy metals Cd, Cr and As were detected, the concentration range was 0.075~0.5mg/kg, 2.24~ 4.09mg /kg, 0.002~ 0.005mg /kg, respectively. The concentration from large to small is: Cr > Cd > As, and the mass

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concentration of Cr accounts for about 90% of the total concentration of the three heavy metals. According to GB/T35601-2017《Green Product Evaluation of wood-based panels and wood Flooring》, the total amount of soluble heavy metals is less than or equal to 100mg/kg, and the total amount of soluble heavy metals in this experiment is between 2.317 and 4.595mg/kg, which is far less than the limit value. See Table 2

Table 2: Average mass concentration of Heavy metals released by laminate flooring at different times (μg/g) (RH50%)

statistical magnitude	Cd mg/kg	Pb mg/kg	Cr mg/kg	As mg/kg
least value	0.075	ND	2.24	0.002
maximum value	0.5	0.012	4.09	0.005
arithmetic average	0.30	0.09	3.92	0.003
deviation	0.142	0.026	0.602	0.001
mid-value	0.113	0.010	2.888	0.113
Geometric mean value	0.180	0.000	3.308	0.003
Degree of variation	28.4	28.4	14.7	21.2

Note: ND indicates not detected

Hu Yunan [8] studied the characteristics and sources of heavy metal pollution in indoor dust fall in Jinan City, and found that Cr was generally slightly polluted, while other elements were generally pollution-freeChen Jing et al. [9] studied that the average concentration of Cr in household indoor dust was 50.20mg/kg, which was the highest concentration among the five metals. Huang Hao et al. concluded that on the whole, Cr in indoor dust was moderately polluted in urban areas and mildly polluted in rural areas. According to source analysis, Cr content is mainly affected by natural sources [10]. Li Zhihao [11] studied that the concentration of Cr in PM2.5 of residential indoor air in Changsha City is at a high level, and Cr has a high content in wood floor finishing layer, which may be gradually released into dust through wood floor wear or leaching.

4.2 Migration of heavy metals in the air

Current studies have shown that [12-13] traffic sources, coal burning, and industrial emissions contribute about 85% to heavy metals in the air and are the main sources of pollution. Printers/copiers, household appliances, furniture accounted for about 10%, decoration, decorative materials accounted for about 3%, 2% for other unknown sources. See Figure 1.

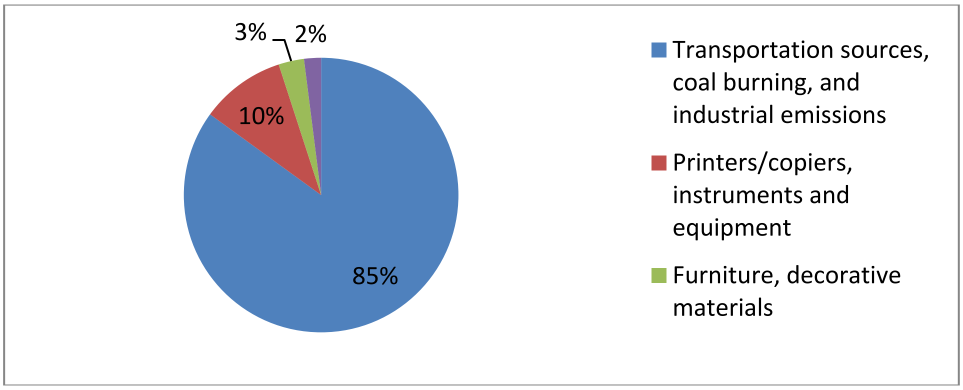

Figure 1: Sources of indoor heavy metals

The sampling season of this study is from February to May, which is the season of high humidity in Yining city. Cr, As and Pb are easily adsorbed by water phase, and the release path of heavy metals from wood floor is complicated. Through the determination of heavy metals in wood flooring, this study found that although the release of heavy metals in wood flooring to indoor air is much smaller than that of the three major pollution sources, it is a long-term and slow release process, which may gradually increase with the wear and tear of wood flooring surface layer, and the content of Pb and Zn in indoor dust will increase with the increase of house age. The material of indoor floor and wood flooring will increase the content of heavy metals Pb and Zn [14]. The release process of heavy metals is affected by PH, temperature, humidity, etc. Due to the limited research data on heavy metals in wood flooring, the release pathways still need to be further studied. It is generally believed that timely cleaning of indoor dust every day will effectively reduce the content of heavy metals such as Cr and Pb

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in the indoor environment and reduce the risk.

4.3 Health risk assessment

Composite wood floor contains four kinds of heavy metals Pb, Cd, Cr, As, in addition to affecting air quality, infants and young children crawling on the wood floor and other behaviors, due to skin contact with the wood floor may have direct health risks. In this paper, the health risk assessment of crawling behavior in three age stages from 0 to 3 years old was studied. The range of crawling activity of infants was about 12 m2. At present, there is no skin absorption data of Pb, Cd, Cr and As. This experiment assumes that all the 4 heavy metals are absorbed by the skin, and the absorption rate is 100%. See Table.3.

According to formula (1) to formula (3), the carcinogenic risk values (ILCR) of As and Cd are 8.4×10^{-4}, 8.4×10^{-6}, respectively, for children 0 to 3 years old. Compared with Table.4 ILCR values at 0 to 1 years old are the largest, which is related to body weight and skin contact area, and there is a certain risk. The carcinogenic risk of As is much lower than that of Cd, which is different from the conclusion of Wang Xianqin [15]. Analyzing the reasons, it is possible that the concentration of As in the wood floor is much smaller than that of Cd, while in the air, the volatility of As is greater than that of Cd, and the concentration of AS entering the human body through breathing is relatively high.

Table 3: Parameter values of respiratory exposure evaluation model

risk type	compound	RfD (mg·(kg·d)−1)	CSFO (mg·(kg·d)$^{-1}$)	0~1 years old	1~2 years old	2~3 years old
carcinogenic	Cd	0.00001	6.1	8.5 x10^{-4}	8.1 x10^{-4}	8.2 x10^{-4}
	Pb	0.000525	/	/	/	/
	Cr	0.00006	/	/	/	/
	As	0.000123	1.5	8.5 x10^{-6}	8.1x10^{-6}	8.2x10^{-6}
Comprehensive				8.6 x10^{-4}	8.6 x10^{-4}	8.3x10^{-4}

Table 4: Chronic oral reference dose and oral slope factor values of heavy meatla

infant	weight /kg	Skin contact area m^2	Floor area m^2	Transfer percentage %	Duration of exposure AT
0~1 years old	8.3	0.11	12	100	25550
1~2 years old	11.2	0.14	12	100	25550
2~3 years old	13.5	0.17	12	100	25550

In this study, heavy metal content in wood flooring was used to calculate the exposure amount, and the risk assessment of human exposure characteristics was carried out. Considering the relative stability of Cr, As and other two heavy metals Cd and Pb in CCA, it was assumed that 100% was adsorbed by the skin, which did not exist in reality, and the normal skin absorption rate was very low, usually only 7% or even lower. Cancer slope factor values are incomplete, making it difficult to accurately assess the health risk of CCA. For such compounds, it is urgent to further study and assess the potential health risks, analyze the natural emission sources of heavy metals, and establish a more complete emission inventory.

5. Conclusions

According to GB/T35601-2017 "Green Product Evaluation of wood-based panels and wood flooring", the wood flooring measured in this experiment did not appear excessive heavy metal phenomenon, cancer risk between 8.1x10-4~8.1x10-6, non-cancer risk HQ<0.1. For enterprises, composite wood flooring enterprises need to strictly check all the production links that may produce heavy metals, and strictly control the heavy metal content within the scope of national standards; Whether it is a national policy or market trend, or consumer demand, the development direction of the industry is safety, health, environmental protection, floor enterprises should pay attention to the measurement standards, take the road of green health and environmental protection.

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