

Variation of arsenic concentration on surfaces of in-service CCA-treated wood planks in a park and its influencing field factors

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Abstract Wood preservatives can protect wood from dry rot, fungi, mould and insect damage, and chromated copper arsenate (CCA) has been used as an inorganic preservative for many years. However, wood treated with CCA has been restricted from residential uses in the EU from June 30, 2004, due to its potential toxicity. Such a regulation is not in place in China yet, and CCA-treated wood is widely used in public parks. A portable XRF analyser was used to investigate arsenic (As) concentration on surfaces of in-service CCA-treated wood planks in a popular park as well as the influencing field factors of age in-service, immersion and human footfall. With a total of 1207 readings, the observed As concentrations varied from below the detection limit (<10 mg/kg) to 15,746 mg/kg with a median of 1160 mg/kg. Strong variation of As concentrations were observed in different wood planks of the same age, on the surface of the same piece of wood, inside the same piece of wood, and different surfaces of walkway

planks, hand rails and poles in the field. The oldest planks exhibited high As concentrations, which was related to its original treatment with high retention of CCA preservative. The effect of immersion in the field for about 4 months was insignificant for As concentration on the surfaces. However, a significant reduction of As was observed for immersion combined with human footfall (wiping by shoes). Human traffic in general caused slightly reduced and more evenly distributed As concentrations on the wood surfaces. The strong variation, slow aging and relatively weak immersion effects found in this study demonstrate that the in-service CCA-treated wood poses potential health risks to the park users, due to easy dermal contact especially when the wood is wet after rainfall. It is suggested that further comprehensive investigations and risk assessments of CCA-treated wood in residential areas in China are needed, and precautionary measures should be considered to

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reduce the potential risks to residents and visitors, especially children.

Keywords Chromated copper arsenate (CCA) · Arsenic · Wood · Portable XRF · Immersion · In-service

Introduction

Chromated copper arsenate (CCA) is a wood preservative containing chromium (Cr), copper (Cu) and arsenic (As). From the 1930s, CCA has been used in pressure-treated wood to protect wood from rotting due to insects (such as termites, borers, beetles) and microbial agents (Hingston et al. 2001; APVMA, 2005; US EPA United States Environmental Protection Agency 2011), thus extending the service time of wood, especially for outdoor uses. It was used to replace previous organic preservatives such as creosote and coal tar due to environmental and health concerns (Hingston et al. 2001).

Even though CCA helps to reduce the use of wood by extending its service time, the negative influences of environmental pollution and potential human health risk have been recognised due to leaching of the agents (Mercer and Frostick 2014; Mercer and Greenway 2014). The main agents of CCA including Cr, Cu and As are listed as priority pollutants by the US Environmental Protection Agency (Weis et al. 1992). Leaching of As from CCA-treated wood has been widely observed in the field (Stilwell and Gorny 1997; Zagury et al. 2003; Khan et al. 2006; Shibata et al. 2007) and in the laboratory (Shibata et al. 2006; Mercer and Frostick 2012) as well as after disposal (Jambeck et al. 2006; Moghaddam and Mulligan 2008). The toxicity of As to human health has been widely recognised, and the potential As-related risk to children playing on CCA-treated wood has been well documented (Dang and Chen 2003). European Commission Directives 2003/2/EC (EC 2003) and 2006/139/EC (EC 2006) have restricted the use of arsenic-treated wood from “residential or domestic constructions, whatever the purpose,” starting from June 30, 2004. Such a limit has also been implemented in other countries including the USA (from December 31, 2003) (US EPA United States Environmental Protection Agency 2011), Canada and Australia (Veterinary Medicines Authority 2005), but not in China as yet.

In a popular nature reserve in China, Jiuzhaigou National Park, fish were found dead in a lake in winter 2008 with no satisfactory explanations until the summer of 2009 when the authors and other experts discovered that the wood used in the park was CCA-treated. A large quantity of CCA-treated wood was piled on the lake side for replacement of the old CCA-treated wood planks, and the toxic As and Cr were leached by melting snow and entered the lake. This accident caused serious concerns about the existence of CCA-treated wood in public parks in China. Besides the potential detrimental impacts to the ecosystem, one of the major concerns with CCA-treated wood in the park was the direct dermal contact of visitors including children who are most likely to exhibit mouthing behaviour (Read 2003), and as expected, As on CCA-treated wood surfaces have been found “dislodgeable” (Stilwell et al. 2003). Therefore, in order to assess the potential health risks to the park users and local residents, it was necessary to investigate As levels on the wood surfaces.

The objectives of this study were to investigate the variability of As concentrations on various surfaces of in-service wood planks and the potential influencing field factors, in order to obtain an understanding potential hazards of CCA-treated wood still in-service in the park as a preliminary step.

Materials and methods

Study area

The study area is Jiuzhaigou National Park, Sichuan Province, Southwest part of China (Fig. 1). It is approximately 640 km² in area with international recognitions of a UNESCO World Heritage site in 1992, the World Biosphere Reserve in 1997 and a “Green Globe 21” certificate in 2001 (Jiuzhaigou National Park 2014). The bedrock of the area is mainly limestone, with transparent water of little known pollution. Wood planks are widely used in the park as the walkways to protect the soils from being stepped on by tourists in the nature reserve. The total length of the wood planks in the park is more than 70 km (Jiuzhaigou National Park 2014). It was regarded as an environmental-friendly measure but little information is available about its “side effects” of potential hazards in the park.

While the wood planks have been constantly replaced and maintained, there are generally three

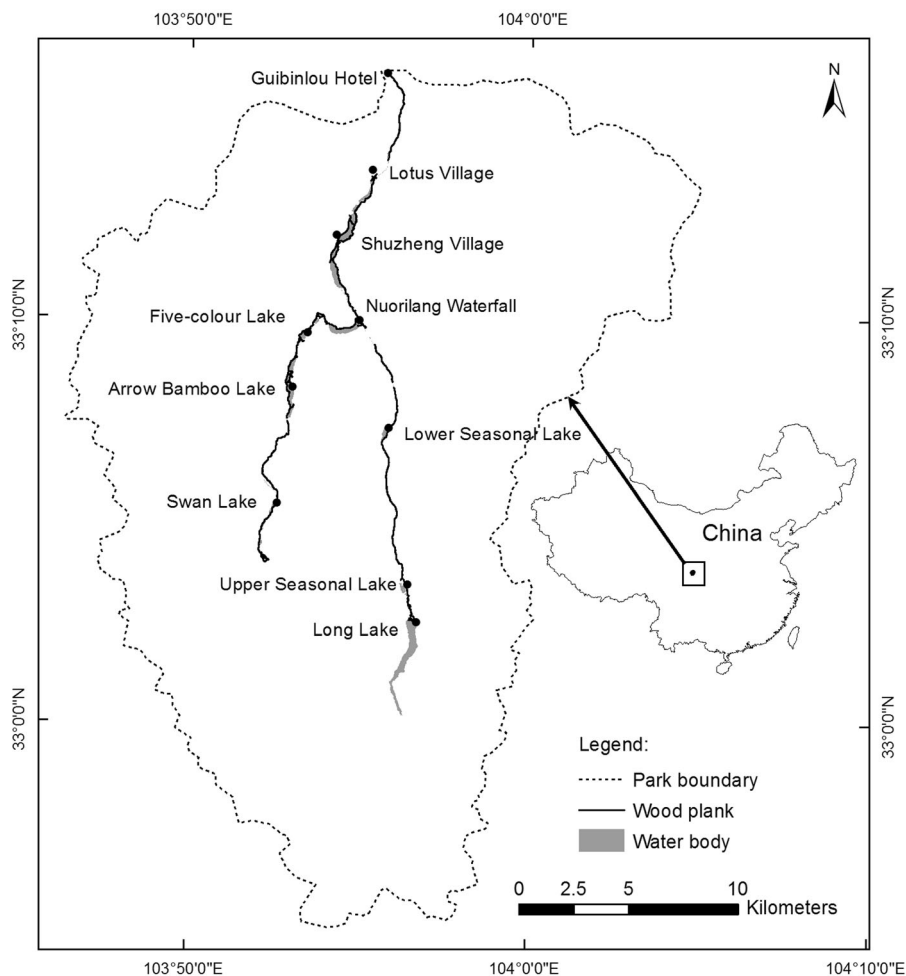


Fig. 1 Study area and distribution of CCA-treated wood planks

generations of wood planks based on the age of the in-service time: The first generation (named as “CCA1”) was installed at least 10 years ago, while the installation of the second-generation wood planks (named as “CCA2”) started in 2006 and was completed in 2012. Both of them were CCA-treated. Following suggestions from the authors, the park started to use alkaline copper quaternary (ACQ)-treated wood planks (the third generation) to replace the old CCA-treated wood from 2013, but the new generation of wood planks was installed in limited areas.

Fieldwork and sampling design

Fieldwork was carried out in July, 2013, using a portable X-ray fluorescence (XRF) analyser (Thermo Niton XL3t). The portable XRF system has been found as a reliable method to measure As on CCA-treated wood

(Blassino et al. 2002; Solo-Gabriele et al. 2004; Yasuda et al. 2006; Block et al. 2007; Jacobi et al. 2007). This equipment detects the As concentration of the surfaces of wood samples, with a detection window of about 1 cm². The readings can be obtained in less than 10 s. In this investigation, the measurement time was set at 60 s in order to obtain reliable results, with the detection limit of 10 mg/kg.

The species of the treated wood used in the park was *Pinus sylvestris var. mongolica* Litv., imported from Russia. Various sampling design methods were employed to investigate the variation of As on surfaces of the treated wood planks as well as influencing field factors, with the details listed in Table 1. A simple random sampling method was applied to investigate variation within each generation of treated wood (with a total of 50 readings on randomly selected wood planks from each generation), variation on a surface of a single

Table 1 Sampling methods for investigation of the variation and influencing field factors of As concentrations on surfaces of treated wood

Generation (<i>n</i>)	Purpose	Sampling method	Description
CCA1 (50) CCA2 (50) ACQ (50)	Variation within each generation	Simple random sampling at each site	CCA1: 50 random readings were made at a site (Arrow Bamboo Lake) where a CCA1 wood walkway was found; CCA2: 50 random readings were made in a warehouse where CCA2 walkway planks were piled; ACQ: 50 random readings were made in a warehouse where ACQ pole planks were piled
CCA1 (100) CCA2 (100) ACQ (100)	Variation within single wood planks	Simple random on each surface	CCA1, CCA2, ACQ: A plank of wood was randomly collected from each generation, and cut into two long pieces in the middle to enable readings on both interior and exterior surfaces. A total of 50 random readings were made on each of the 6 surfaces
CCA1 (110) CCA2 (350)	Variation among different surfaces of planks; comparison between generations (CCA1 and CCA2)	Cluster sampling at various sites	CCA1: Multiple random readings were made at each of 8 sites where CCA1 wood was found. The sampling parts included walkway top (<i>n</i> =40), side (30) and end (40). Handrails and poles were not found for CCA1 wood planks. CCA2: Multiple random readings were made at each of 12 sites where CCA2 wood was found. The sampling parts included walkway top (49), side (43) and end (43); handrail top (43), side (43) and bottom (43); pole top (43) and side (43)
CCA2 (210) CCA2 (53)	Influences of immersion	Simple random sampling at a site; systematic sampling on steps	Multiple random readings were made at Long Lake site where CCA2 wood was immersed in water during May–Sept., 2012. The sampling surfaces included walkway top (30) and end (30); handrail top (30), side (30) and bottom (30); pole top (30) and side (30). It was problematic to take readings on walkway side (0). Multiple random readings (at least 5) were made on each step of 10 steps from the bottom of a stair where the planks was immersed in water in summer 2012 at Long Lake site. A water mark was observed on the 8th step
CCA1 (184)	Influences of human footfall	Systematic sampling on a walkway plank	Outside Guibinlou Hotel, a total of 3 obviously stepped on planks and 3 clearly un-stepped planks (located immediately below the edge of a step) were randomly chosen for measurements at an interval of 5 cm from end to end of each plank

wood plank and influences of immersion. Cluster sampling was employed to compare the As concentrations on different surfaces and between different generations. Finally, the systematic sampling was used to reveal the influences of water immersion on steps (with different heights) and the influences of human footfall (distance from end to end on a single wood plank).

Data analyses

Raw data were exported from the portable XRF analyser to an MS Excel file. Basic statistics were calculated using MS Excel and IBM SPSS (Ver. 20). SPSS was applied to perform statistical analyses and produce

charts. Quality control was achieved using a reference sample RCRA STD (part number 180-436, Thermo Scientific) measured in the field. The recovery rate varied from 98 to 114 %, with a median of 104 %. The results for untreated wood were below the detection limit of 10 mg/kg.

Results and discussion

Variation within generation

It is hypothesised that different pieces of wood planks from the same generation may not contain the same

amount of As on their surfaces, and the results are shown in Table 2. Strong variation of As was observed in different planks of the same generation/batch of treated wood, showing the strong heterogeneity of wood treatment. While As concentration in ACQ-treated wood was at a negligible level, it varied from <10 mg/kg to several hundred or thousand milligrams per kilogram in both CCA1 and CCA2. It should be noted that there were a few readings for ACQ slightly higher than the detection limit which were likely analytical errors but they were close to the detection limit. By chance, the site selected for CCA1 variation analysis contained relatively low As concentrations. It is acknowledged the site for CCA1 was not representative enough, but the variation was strong with a coefficient of variation (CV) of 163.7 %. The variation based on readings from multiple sites showed similarly strong heterogeneity of As concentrations within the same generation, with the maximum values of As for both CCA1 and CCA2 higher than 1 %. With a total of 1207 readings for CCA-treated wood in this study (1164 above the detection limit), the observed As concentrations varied from <10 to 15,746.1 mg/kg with a median of 1159.9 mg/kg. The very low values for the CCA-treated wood could be caused by various factors including degradation, sharpening (to create the cone shape of the top) and human footfall as well as the natural variation.

Variation on the same surface of a single wood plank

To investigate the variation of As concentration on the same surface, a total of 50 readings were made on each exterior and interior surface of a wood plank from each generation. Heterogeneity of As was observed on both surfaces of the same plank of wood (Fig. 2). This was clear for both CCA1 and CCA2, while the results for ACQ were generally below the detection limit as expected. It was noted that the variation on the same

surfaces was generally within similar magnitudes, which was much smaller than that within a generation.

For CCA1, As concentrations inside the wood plank were significantly higher than those on the exterior surface (Mann-Whitney *U* test $p < 0.05$), with the median values of 11,476.6 mg/kg, in comparison with the median of 10,617.4 mg/kg on the exterior surface. Such a result demonstrated that the preservative had been distributed through the wood, and the exterior surface exhibited the effects of over 10 years of leaching in the field.

As for CCA2, the interior As concentrations were much lower than those on the exterior surface (Mann-Whitney *U* test $p < 0.05$). The medians were 59.8 mg/kg for the interior surface and 1385.8 mg/kg for the exterior surface, respectively. It demonstrated that CCA agents were mainly distributed on the surfaces, and the treatment did not distribute much CCA inside the wood. While such an observation was based on only one wood plank, the difference between the interior and exterior surfaces was apparent. In terms of CCA treatment, the quality of CCA2 was possibly “poorer” than that of CCA1 with respect to concentrations of As.

Variation on different surfaces of wood planks

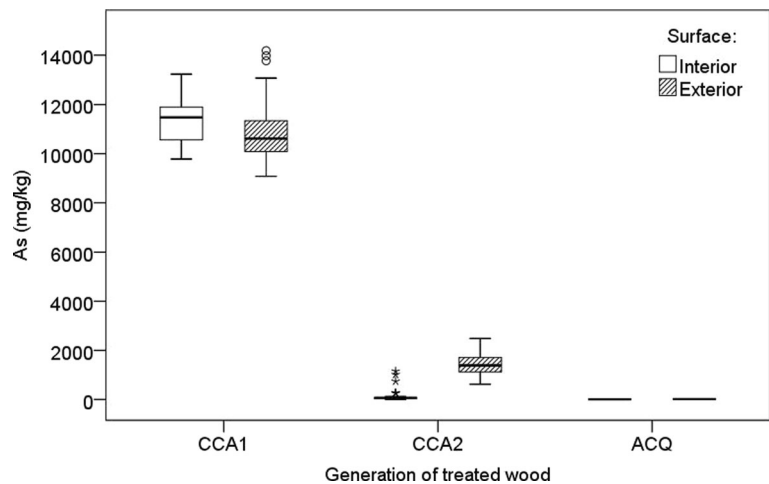
CCA-treated wood planks were used as walkways, hand rails and poles in the park. These three types of usages were widely found for CCA2. Therefore, CCA2 was chosen to investigate the possible variation of As concentration on these different usages. Different surfaces were considered as follows: top, side and end, except for the end surfaces of poles as only pole top and side surfaces were possible to measure in the field. A total of 43 readings were made for each of these surfaces, and 49 readings for walkway top surfaces (Table 1).

As expected, strong variation of As concentrations were observed on all the surfaces of various plank

Table 2 Variation of As concentration in different planks of the same generation of treated wood (in mg/kg)

Sampling site	Number	Generation	Min.	25 %	Median	75 %	Max.	Mean	StdDev	C.V. (%)
One pile	50	ACQ	<10	<10	<10	<10	14.2	–	–	–
One site	50	CCA1	<10	14.2	20.1	28.9	371.0	33.6	55.0	163.7
One pile	50	CCA2	81.1	517.4	1767.3	3788.5	8505.7	2546.1	2343.8	92.1
8 sites	110	CCA1	<10	509.7	2246.7	3964.2	14,003.0	2733.9	2771.0	101.4
12 sites	350	CCA2	<10	408.9	1243.9	2815.4	15,746.1	2014.0	2286.3	113.5
All readings	1207	CCA1+2	<10	364.0	1159.9	2678.4	15,746.1	2382.1	3218.2	135.1

Fig. 2 Variation of As concentration on the surfaces of a single plank of treated wood ($n=50$ for each surface)

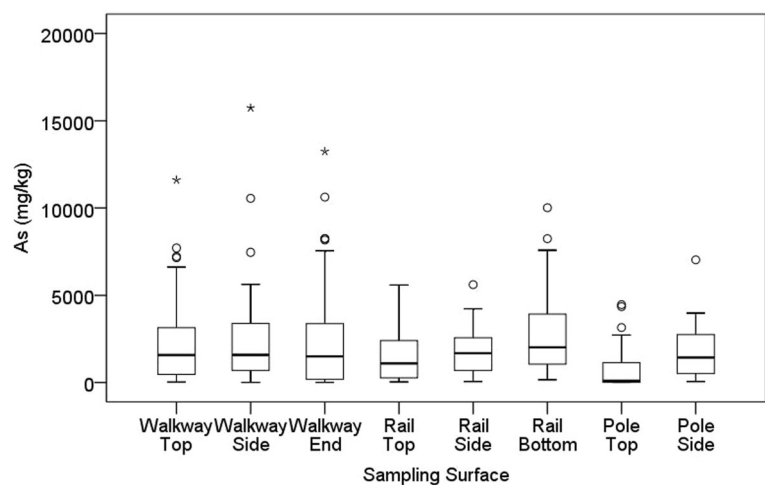


usages (Fig. 3). The ranges varied from <10 mg/kg to several thousand milligrams per kilogram, with six values even higher than 1 %. However, apart from the pole tops, the median values of the surfaces were similar, with insignificant differences for both median (median test $p>0.05$) and distribution (Kruskal-Wallis test $p>0.05$). When pole tops were included, the differences between the surfaces for both median and distribution tests were significant ($p<0.01$). The special behaviour of pole top should be related to a process prior to installation: the surfaces on the top ends of the poles were removed and sharpened to create conic shapes. As observed earlier, the interior of CCA2 contained low concentrations of As due to the possible “poor” CCA treatment in the factory. The sharpening operation of the pole tops exposed the interior part of wood to the surface, and the surfaces were unevenly sharpened,

which could be one of the reasons causing the strong variation of As concentrations. Such an observation also re-confirmed the possibly poor CCA treatment in the factory for CCA2.

Apart from the pole top (median 104.8 mg/kg), the other surfaces shared similar features of As concentrations, e.g., strong variation, similar medians (from 1093.5 mg/kg for rail top to 2017.6.5 for rail bottom) and distribution. The slightly higher As concentrations on rail bottom surfaces could be partly related to leaching but its differences with the other surfaces (excluding pole top) were insignificant. In general, strong variations were observed for all surfaces of planks used as walkways, hand rails and poles. However, there were no significant differences between them, except for the relatively low As concentrations on the pole tops due to the sharpening operation.

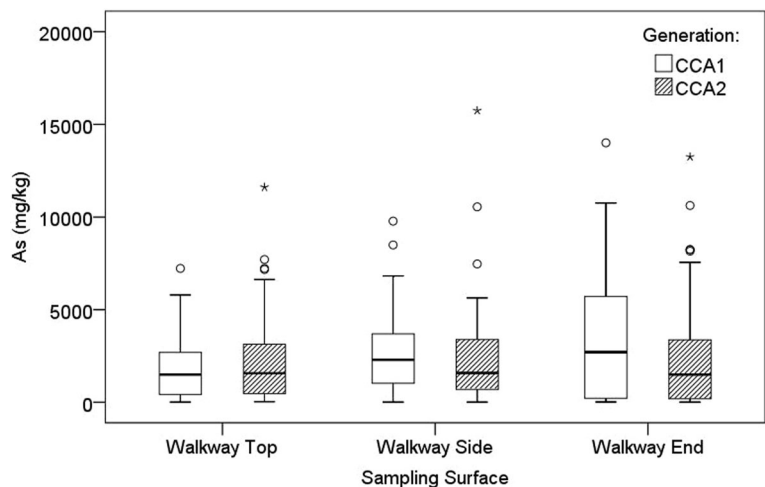
Fig. 3 As concentrations on different surfaces of wood planks (CCA2)



Difference in generations

Time has been recognised as an important factor affecting As concentrations on surfaces of CCA-treated wood (Brooks 1996). It is reasonable to propose that the longer the treated wood planks in service in the field, the more As could be leached, causing possible reduced As concentrations on the surfaces. Comparisons between corresponding surfaces of CCA1 and CCA2 were made based on multiple readings in the field (Fig. 4). The result was beyond expectation: As concentrations on all three surfaces (top, side and end) of CCA1, with much longer time in the field, were similar to those on CCA2 with insignificant differences (Mann-Whitney *U* test $p > 0.05$ for all three surfaces). The median values for side and end of CCA1 were even slightly higher than those of CCA2, implying different processes of preservative in the factories: CCA1 was possibly “better” processed than CCA2 in terms of the use of preservatives, as observed earlier in this study. Meanwhile, the leaching effect on As concentrations on the surfaces was slow and limited, especially with the increase of time in service. A recent study by Coudert et al. (2014) revealed that the As leaching with aggressive conditions became more difficult with the increasing of the elapsed time between wood preservation and the decontamination. They explained that it could be due to the fact that the As that was not well fixed to the wood during the impregnation was leached during the first years of wood service time and that the remaining As was strongly fixed to the intrinsic components of wood.

Fig. 4 Comparison of As in different generations of treated wood: CCA1 and CCA2

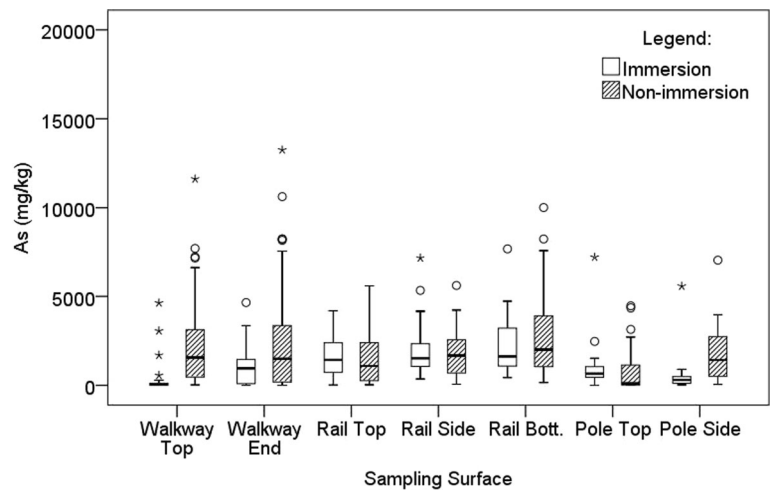


Influences of immersion in the field

During May to September, 2012, the CCA2 planks along the side of Long Lake in the upper part of the park were immersed in water for four months due to heavy rain. The potential influence of immersion on As concentrations were investigated using a comparison with corresponding plank surfaces of the other CCA2 without immersion (non-immersion) in the park (Fig. 5).

Clear differences between immersion and non-immersion samples were identified on surfaces of walkway top and pole side, which were confirmed by the Mann-Whitney *U* test ($p < 0.05$). The differences between immersion and non-immersion samples on other surfaces were insignificant ($p > 0.05$), except for pole top ($p < 0.05$). The most significant influence of immersion was found on walkway tops where the majority of the immersion samples were below or close to the detection limit (10 mg/kg). This could be related to the constant human footfall while the walkways were wet. CCA agents could have been “wiped” away by shoes of tourists. The relatively lower As concentrations on the immersed pole side surfaces could be related to the “washing” by water when the planks were in the vertical position. The relatively high As concentration on the immersed pole tops was related to the flat shape of the tops along the Long Lake. Unlike the cone-shaped tops, these tops were only slightly cut on the four edges prior to installation. Overall, the influence of immersion on As concentrations of plank surfaces during the summer 2012 was generally insignificant under the slightly alkaline conditions (pH value around 8 based on unpublished monitoring results from the park). However, the

Fig. 5 Comparison between As concentrations in treated wood surfaces (CCA2) with and without immersion in the field



combined effect with human influences of constant “wiping” by shoes was obvious.

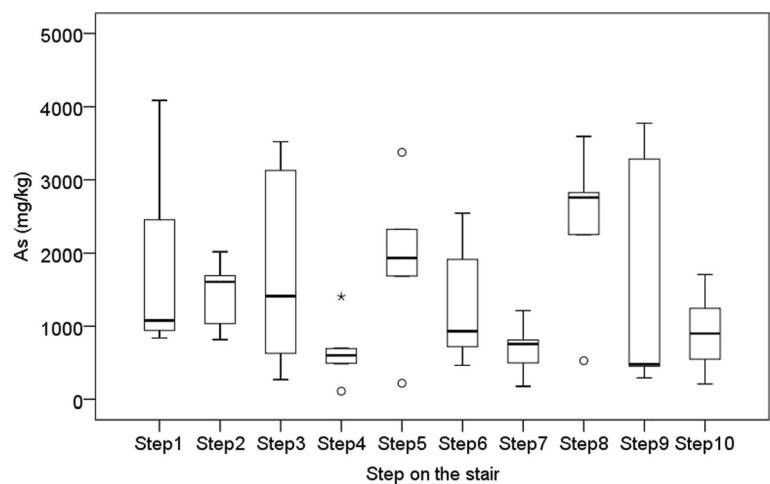
A further investigation on the immersion effects was made on variation of As concentrations on different steps of a plank stair located near the Long Lake. The assumption was that the lower steps were immersed in water longer than the higher steps; thus, it was reasonable to expect variations in As concentration on different steps. Starting from bottom to top (with the highest water mark of the 2012 summer immersion event on step 8), the steps on the stair were numbered from 1 to 10 (Fig. 6). No clear trend of variations on the different steps was found even though multiple readings were made on each step for a more robust result. The random readings (at least 5) made on each step varied strongly between 109.6 to 5087.0 mg/kg. The strong variation of

As concentrations may have also contributed to such a result.

Influences of human footfall

The final field factor investigated in this study was human footfall. Since the wood planks were constantly stepped on by park visitors and residents, a site immediately outside the park gate leading to a popular hotel was chosen. The walkway was about 100 m long, with a mild slope. The CCA1 planks along the walkway stair were installed in multiple steps, and each step contained several horizontal planks. The planks immediately under the steps were unlikely stepped on, and the third planks from the upper step edge were well stepped on based on visual identification. Three non-stepping and

Fig. 6 Variation of As concentrations on a stair affected by immersion



other three stepped on planks were chosen for analysis. Arsenic concentration on each plank was measured from one end to the other end at an interval of 5 cm (Fig. 7).

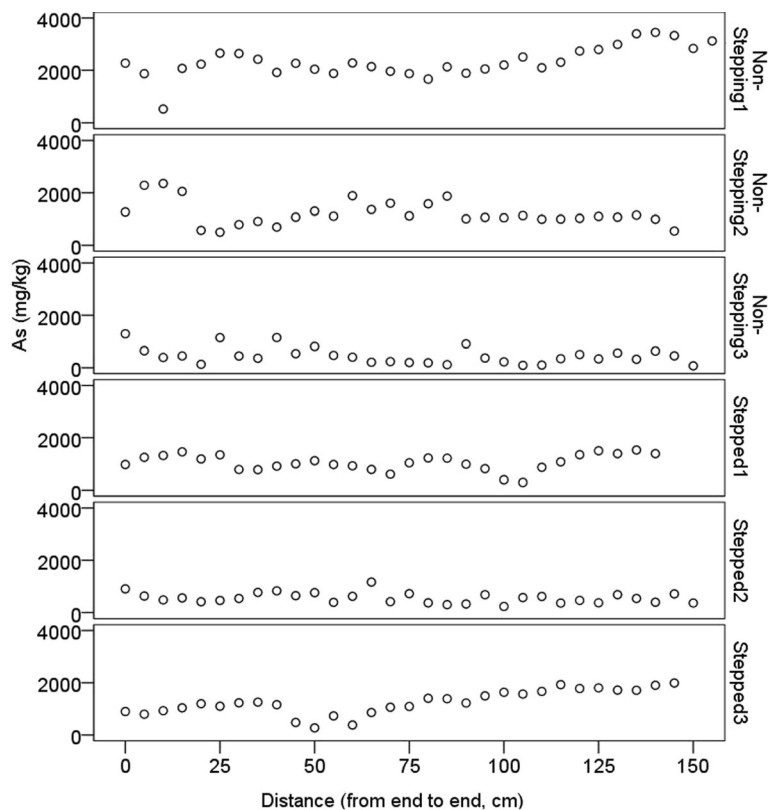
As expected, strong variations of As concentrations on all the six planks were observed, especially on the planks which were not stepped on. The stepped-on planks generally showed slightly lower As concentrations than the non-stepping planks. For the stepped-on planks, generally lower As concentrations were found in the middle part, showing some level of influences of footfall. The variation along the plank was relatively smooth on the stepped-on planks implying the “mixing” or “smoothing” effect of human footfall. The relatively weak footfall effect on CCA1 could also be related to the fact that As distributed throughout the whole CCA1 planks as observed earlier. Further studies can be considered on the footfall effects on different generations of CCA-treated wood and different locations.

Finally, it needs to be acknowledged that this study was based on field investigation only. The field influencing factors are complicated, and the field

conditions are not well under control as those in a laboratory. Results obtained in this study were made in one park only, but it is expected that such results could be applicable elsewhere. This is the first such study in the park, and the results revealed high concentrations of As on all surfaces of CCA-treated wood planks still in-service. The potential health effects on the residents, park staff and tourists remain unknown.

Since CCA-treated wood is not restricted for domestic use in China yet, the CCA-treated wood is still widely used in tourist destinations. More studies should be carried out on risk assessments to establish enough evidence to legislate on the use of CCA-treated wood for domestic use as in the EU and many other countries. In the absence of such legislations in China, precautionary measures should be taken. Tourists should avoid direct dermal contact with the CCA-treated wood especially when it is wet after rainfall. The park management should ensure that once the CCA-treated wood is out of service, it is classified as hazardous waste and thus should be properly disposed of. For residents, the strong message of “no burning” should be passed to them as

Fig. 7 Distribution of As concentrations along walkway planks from end to end (three planks unlikely to be stepped on, and three planks clearly stepped on)



many of them may not be aware of the risks of high As concentrations in such wood.

Conclusion

A portable XRF analyser was found useful to measure As concentration on surfaces of CCA-treated wood planks in the field. Strong variations of As concentration on surfaces of wood planks in the park were observed. Such variations of As concentration were observed among different wood planks of the same age/batch of treatment, on the surface of the same piece of wood, inside the same piece of wood and different surfaces of walkway planks, hand rails and poles in the field. The planks of the oldest generation showed similar or even slightly higher As concentrations than the newer generation, showing its original treatment with possible high concentrations of CCA agents. The newer generation of CCA-treated wood also exhibited lower As concentrations on the interior surfaces, demonstrating its possible poor treatment in the factory with respect to concentrations of As. The effect of immersion in the field for about 4 months was found insignificant on As concentration, but it significantly reduced As concentrations on walkway planks in combination with constant human footfall. Human footfall slightly reduced As concentrations on wood planks, and also showed a smoothing effect on the distribution of As concentration on the stepped on wood surfaces. Further comprehensive investigations and risk assessments of CCA-treated wood in residential areas in China are needed, and precautionary measures should be considered to reduce the risks to residents especially children.

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