Biological Leaching of Metals from Respirable Tire Wear Particles

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ABSTRACT

Respirable particulate matter (PM_{10}) in the environment has been associated with a number of human diseases. Tire wear particles (TWP), produced from the interaction of the pavement and the tire surface, are thought to contribute to PM_{10}; however, airborne TWP has not been well characterized and thus associated risks are not well understood. In order to address this question, we collected PM_{10} samples while running passenger vehicle tires on a roadway simulator, thus simulating environmentally relevant driving conditions during TWP collection. PM_{10} samples were collected on mixed cellulose ester filters (MCE) and subjected to biological leaching experiments in simulated lung fluid (SLF) in order to understand bioaccessibility of metals found in TWP. The filters were incubated in SLF at 37°C for 6 days under constant agitation at 80 rpm. Leachate from the PM10 air samples were analyzed for metals content by ICP.

INTRODUCTION

For the past 15 to 20 years, there has been a significant amount of research conducted examining the relationship between ambient particulate matter (PM) and human health effects. Source apportionment studies indicate that motor vehicle emissions are a significant contributor to ambient PM; and although the majority of the research has focused on tailpipe emissions, some studies have suggested that non-exhaust sources, such as tire wear, may contribute to ambient PM. Tire wear particles (TWP) result from the interaction of tires with the road surface, and include both the rubber matrix and any embedded minerals from the road. TWP are a component of road dust (Rogge et al. 1993; Camatini et al. 2001; Kreider et al. Submitted April 2009). Inhalation of tire-containing particles has been associated with inflammation, cytotoxicity, and genotoxicity (Gualtieri et al. 2005; Mantecca et al. 2009); however, these studies used organic extractions and exposure conditions that are not representative of real-world exposure. Soluble transition metals, such as zinc and copper, from tire particles have been implicated in a number of studies as causative of pulmonary toxicity (Gottipolu et al. 2008; Kodavanti et al. 2008). In our previous work, we collected TWP at a road simulator which eliminated influence from other vehicle particulate (exhaust, brake dust, etc.). We have previously presented physical/chemical characterization, acute aquatic toxicity and environmental leaching of TWP. In the leaching study, we found that only zinc leached from TWP under environmental leaching conditions (McAtee et al., in preparation). Therefore, the goal of this work was to characterize metals in simulated lung leachate following TWP incubation.

METHODS

TWP was collected at a roadway simulator as previously described (Figure 1; Kreider 2009). Passenger tires including summer and winter silica tires and carbon black based tires were utilized. PM_{10} was collected for 40 hours using PM_{10} samplers at a flow rate of 16.7 lpm. A total of ten PM_{10} samples were collected for biological leaching experiments. Based on previous work, the filters were analyzed for metals content. Therefore, mixed cellulose ester (MCE) filters, which meet NIOSH and OSHA specifications for airborne metal monitoring and are low in background metals, were utilized. In order to increase the PM mass for leaching, three filters were used in each leachate experiment. Three separate leaching experiments were conducted (n=3). The filters were incubated in simulated lung fluid (SLF; Table I). High purity salts were used to produce the SLF in order to reduce levels of background metals. The SLF (pH 7.2) was prepared and used under sterile conditions at all times. The SLF was infused with, and capped in, an atmosphere of 5 vol % CO_2 in O_2 to maintain the pH. The pH was checked daily, adjusted back to the original pH with dilute HCl as needed and then regassed (Twining et al. 2005; Schauer et al. 2006).

Table 1. Simulated Lung Fluid Recipe*

Salt*	Formula	Concentration (mM)	MW	Mass (g/L)
Sodium chloride	NaCl	110	58.4	6.43
Sodium bicarbonate	$NaHCO_3$	31	84.0	2.60
Calcium acetate	$Ca(C_2H_3O_2)_2$	2.5	158.2	0.40
Calcium chloride	$CaCl_2 \cdot 2H_2O$	2.5	147.0	0.37
Magnesium acetate	$Mg(C_2H_2O_2)_2 \cdot 4H_2O$	1	214.5	0.21
Magnesium chloride	$MgCl_2 \cdot 6H_2O$	1	203.3	0.20
Potassium dihydrogen phosphate	KH_2PO_4	2	136.1	0.27
Dipotassium sulfate	K_2SO_4	1	174.3	0.17
Citric acid	$C_6H_8O_7$	1	70.0	0.07
Albumin (fraction 5)		---	---	0.20
Benzalkonium chloride (preservative)		---	---	0.05

*Twining et al., 2005

SLF was passed through Chelex resin column twice to remove trace metals. The MCE filters were placed into clean polypropylene screw-top centrifuge tubes and were only handled with Teflon coated forceps. Blank filters (3 tubes) and SLF-alone (3 tubes) were also analyzed as controls. 25mls of SLF (after Chelex resin treatment) were then added to each tube. The tubes were capped, placed in a 37°C oven and shaken for 6 days at 80 rpm, as 6 days is the maximum residence time of most particles in the lung (Twining et al. 2005; Schauer et al. 2006). At the end of the incubation, the leachate and solid residues were separated using a Puradisc syringe filter (GE Whatman). The final pH of the leachate was recorded and the leachate was then acidified with 3 vol % concentrated analytical grade nitric acid to ensure that the constituents remain in solution. The sample was ashed and dissolved in acid for ICP analysis on a Spectro FTP-08 ICP.

Pavement samples were collected and analyzed via PIXE at the Division of Nuclear Physics at Lund University. The PIXE analysis was performed at the accelerator laboratory using a 2.55 MeV proton beam produced by a single-ended electrostatic NEC accelerator. The x-ray spectra were fitted with the GeoPIXE II program.

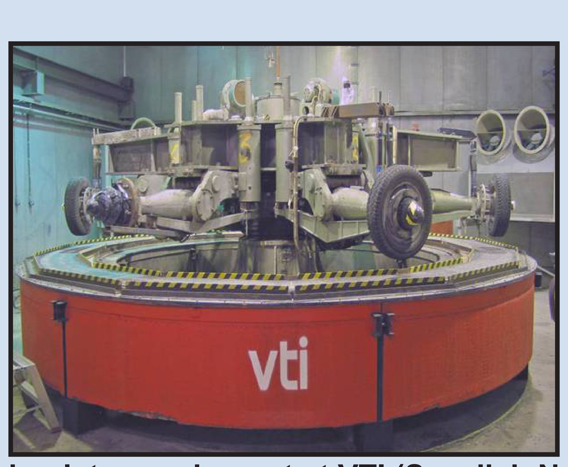

Figure 1. Road simulator equipment at VTI (Swedish National Road and Transport Research Institute) in Linkoping, Sweden.

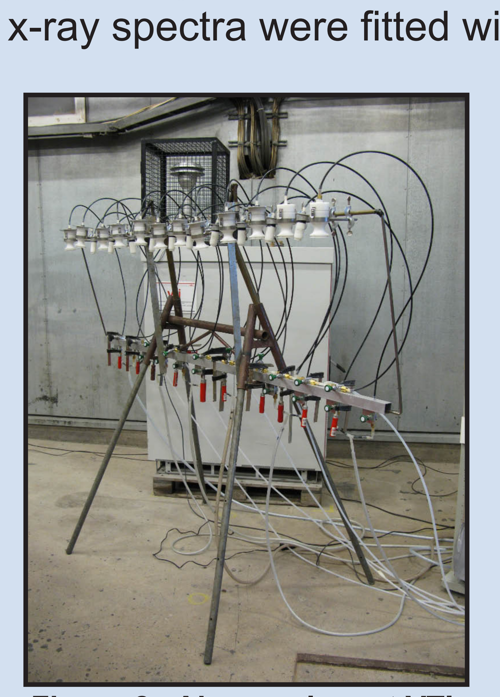

Figure 2. Air samplers at VTI.

RESULTS

Simulated lung fluid leachate from the PM_{10} air samples contained significantly more aluminum and zinc compared to leachate of blank filters ($p<0.05$; Table 2). In our previous work, TWP and tire tread were subjected to elemental analysis by ICP (Kreider et al. Submitted April 2009). From the previous study, we know the concentration of Al, Ca, and Zn in TWP and tire tread (Table 3). The pavement is the source of aluminum, as Al is detected at high levels in pavement samples (Table 3). Tire tread contains zinc oxide (~0.9%) and therefore, it is likely that the tread contributed to the zinc found in the SLF. The total PM10 collected contained an average of 3-5% zinc; therefore 54.5% of the zinc in the PM10 sample was calculated to be bioaccessible in the simulated lung fluid (Table 4).

Table 2. Metals detected in simulated lung leachate by ICP

	TWP PM_{10} R-1	TWP PM_{10} R-2	TWP PM_{10} R-3	Blank (blank filter in SLF)
Aluminum, ppm	0.0396	0.0598	0.0619	0.0361
Antimony, ppm	0.091	0.086	0.1089	0.077
Arsenic, ppm	0.2337	0.173	<0.0999	0.2214
Barium, ppm	0.0414	0.0061	<0.0017	<0.0017
Beryllium, ppm	<0.0003	<0.0003	<0.0003	<0.0003
Bismuth, ppm	<0.1212	<0.1212	<0.1212	<0.1212
Boron, ppm	0.1672	0.1624	0.2775	0.1145
Cadmium, ppm	<0.00525	<0.0022	<0.0022	<0.0022
Chromium, ppm	<0.0131	<0.0131	<0.0131	<0.0131
Cobalt, ppm	<0.1293	<0.1293	<0.1293	<0.1293
Copper, ppm	<0.0094	<0.0094	0.0756	<0.0094
Iron, ppm	<0.0294	<0.0294	<0.0294	<0.0294
Lead, ppm	<0.0131	<0.0131	<0.0131	<0.0131
Manganese, ppm	<0.0041	<0.0041	<0.0041	<0.0041
Nickel, ppm	<0.0092	<0.0092	<0.0092	<0.0092
Selenium, ppm	<0.1383	<0.1383	<0.1383	<0.1383
Silicon, ppm	0.4153	0.1727	0.1563	0.99
Silver, ppm	<0.0057	<0.0057	<0.0057	<0.0057
Thallium, ppm	<0.4249	<0.4249	<0.4249	<0.4249
Titanium, ppm	<0.0020	<0.0020	<0.0020	<0.0020
Vanadium, ppm	<0.0314	<0.0314	<0.0314	<0.0314
Zinc, ppm	0.1477	0.1788	0.285	0.0262

Table 3. Concentrations of Al and Zn in tire tread, tire-wear particles, and pavement

Element	Tire tread (ppm)[a]	TWP (ppm)[a]	Pavement (PIXE) (ppm)	
			Stone	Bitumen
Al	470	28,200	ND[b]	ND[b]
Zn	9000	3000	31-168	37

[a]Adopted from Kreider et al., 2009
[b]Al cannot be detected using PIXE

Table 4. Bioaccessible zinc in PM_{10}

TWP PM_{10} sample	Concentration of Zn (ppm)	Volume of leachate (mL)	Mass of Zn (μg)	Corrected mass (-blank) (μg)	Expected mass of Zn (μg)	% Leached
TWP R-1	0.15	40	5.91	4.86	13.03	37.31
TWP R-2	0.18	40	7.15	6.10	13.03	46.85
TWP R-3	0.29	40	11.40	10.35	13.03	79.46
Blank filter	0.03	40	1.05	0	13.03	0.00
					Mean	54.54

DISCUSSION

Zinc oxide, which is used in tires as a vulcanization accelerator, is measured in tread rubber at about 0.9% (9000 ppm) (Table 3) (Kreider et al. Submitted April 2009). In this study, three metals leached from TWP in SLF. While aluminum likely originates from the pavement, the tires are the likely source of the zinc. Bioaccessible zinc was calculated to be 54.5% of the total zinc in the PM_{10} sample. These results indicate that aluminum and zinc may be leached into the lung fluid if/when TWP are inhaled; however, further study is required to fully understand the bioavailability and potential toxicity of these metals.

Inhalation of tire-containing particles has been associated with inflammation, cytotoxicity, and genotoxicity (Gualtieri et al. 2005; Gottipolu et al. 2008; Mantecca et al. 2009); however, these studies used organic extractions and exposure conditions, such as instillation, that are not representative of real-world exposure. These limitations, such as use of organic extractions that would not occur in biological systems, and exposure via instillation which bypasses many of the natural biological defense systems, limit the applicability of these results to human health risk assessment. Potential health effects of inhalation of TWP will require further study.

CONCLUSION

This study found that aluminum and zinc leach from TWP into SLF following incubation for 6 days at 37˚C. Aluminum likely originates from the pavement while zinc likely originates from the tire.

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REFERENCES

Available upon request.