

The environmental fate of traffic-derived metals in a
section of Wet Tropics World Heritage Area
(WTWHA), Far North Queensland (FNQ)

Thesis submitted by:

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Abstract

The major aim of this research was to resolve the following question:

What are the key processes affecting the concentrations, mobility and bioavailability of traffic-derived metals (Cd, Cu, Pb, Ni, Zn, Pd and Pt) in roadside environments in a section of the wet-dry tropics in northern Australia?

Specific areas investigated included the Kuranda Range Road, northwest of the city of Cairns; the Captain Cook Highway at the base of the Kuranda Range Road; and adjoining streams and grassed fields. The Kuranda Range Road traverses World Heritage-listed rainforest and the Queensland Department of Main Roads plans to upgrade the road from two lanes to four.

Materials analysed in the study comprised bedrock, road sediments, road runoff waters, stream sediments, roadside topsoils, and grasses. Additionally, background stream sediment, stream water, topsoil and grass samples were collected away from roads.

Geochemical analyses of the road sediments from the Kuranda Range Road revealed variable total metal concentrations (median values: 0.19 mg/kg Cd, 41.7 mg/kg Cu, 53.3 mg/kg Pb, 38.8 mg/kg Ni, 852 mg/kg Zn, 0.035 mg/kg Pd, 0.086 mg/kg Pt). Moreover, the studied road sediments exhibited metal enrichment (Ni excepted) relative to background stream sediments (maximum enrichment factors: Cd 1.8x, Cu 1.5x, Pb 6.8x, Zn 17.3x, Pd 49.5x, Pt 82x). Partial (citrate dithionite) and sequential (as per the method of Tessier et al. 1979) extractions were performed on the road sediments to examine their metal host sites. The results demonstrated that approximately 35 % to 95 % of the sediments' metal content was accommodated by acid (HF-HNO₃-HClO₄)-insoluble fractions, likely residual silicates. However, significant ($p < 0.01$) positive correlations between the C_{org} and Cd, Cu, Pb and Zn concentrations in the road sediments pointed to metal hosting by an organic source, most likely tyre rubber shreds. The extraction techniques revealed that metals associated with tyre rubber are not readily removed by extraction reagents. Hence, other methods, including correlation analyses between metal concentrations and Al, Mn, Fe and C_{org} values, are necessary to accurately interpret metal hosting within road sediments.

Chemical analyses were performed to evaluate the mobility of Cd, Cu, Pb, Ni and Zn within road runoff waters on the Kuranda Range Road. Maximum Cu, Pb and Zn levels in filtered (<0.45 µm) road runoff waters taken in November 2004 (after a prolonged absence of rainfall) were 8x, 6x and 12x greater than their respective highest values in samples acquired in February 2003 and January 2004 (following heavy rainfall). Such temporal metal distribution data for road runoff waters suggest that large volumes of rainfall in wet-dry tropical regions are capable of mobilising high levels of metals from road surfaces during the initial flushing event (i.e. the ‘first flush’). Furthermore, laboratory leaching and ponding experiments conducted on road sediments indicated that a small proportion of the total heavy metal content (<10 %) of road sediments is readily dissolved in distilled water. In the leach tests, aqueous Cd, Cu, Ni and Zn concentrations showed a pronounced ‘first flush’ effect (i.e. metal values were much higher in the first few samples than in the remaining leachates).

To explore the dispersion of metals from road surfaces, stream sediments from Avondale Creek (intersecting the Kuranda Range Road) were analysed for their total metal contents and Pb isotopic ratios ($^{208}\text{Pb}/^{206}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$). The results revealed: a) elevated total Pb (29.6 mg/kg) and Pt (0.025 mg/kg) concentrations in the sediments collected downstream of the road compared to sediments upstream of the road (Pb = 7.3 mg/kg, Pt = 0.006 mg/kg); and b) non-radiogenic Pb isotopic signatures (characteristic of Broken Hill Pb used in petrol) in sediment samples downstream of roads relative to background stream sediments. The results likely reflect contamination of the catchment by road sources.

The verification of metal contamination within Avondale Creek triggered an investigation into the bioavailability of traffic-derived metals. This involved an assessment of the uptake of soil-hosted metals by a grass species (*Melinis repens*), growing adjacent to the Kuranda Range Road. Median total metal concentrations in topsoils collected adjacent to the road were much higher than median total metal values in topsoils taken 5 metres from the road edge. In the *M. repens* grass specimens, Cu, Pb, Ni and especially Zn concentrations were elevated in roots acquired from immediately adjacent to the Kuranda Range Road. *M. repens* clearly has the ability to incorporate high concentrations of trace elements when growing

on contaminated roadside soils, particularly Zn and to a lesser degree Cu, Pb and Ni. Additionally, extractions using a DTPA-CaCl₂-TEA-HCl (DTPA) solution revealed a significant positive correlation ($p < 0.01$) between soil-DTPA and root Zn levels in the roadside *M. repens* samples. This indicates that the DTPA reagent is a rudimentary indicator of Zn to the roots of this grass species. Metal concentrations in *M. repens* samples grown in road sediments as part of a greenhouse experiment, were similar to the values exhibited by the field specimens. Moreover, the metal levels extracted from the road sediments by an EDTA-NH₄HCO₃ solution were commensurate with DTPA-extractable values, indicating that both of these solutions target similar metal fractions in road sediments.

The final research phase examined remediation measures for road runoff waters on the planned Kuranda Range Road Upgrade. A treatment selection process identified dissolved metals as the most significant category of pollutants because of their high lability and potential toxicity. Site constraints, including the close proximity of the road to sensitive water catchments, indicated that at-source pollutant attenuation will be the most effective remediation option for the road upgrade. Thus, existing at-source primary treatment measures (e.g. trash racks); secondary technologies (including sand filters); and tertiary structures (such as biofilters) were identified as the most suitable treatment options for the Kuranda Range Road Upgrade. Few tertiary treatment devices exist for road runoff waters (the StormFilter is an exception). Hence, this research explored the capacity of commonly-available materials, including mushroom compost and bentonite, to remove dissolved metals from road sediment leachates. In laboratory experiments conducted in this project, mushroom compost and bentonite displayed strong capacities to reduce dissolved heavy metal concentrations in road sediment leachates (Pb and Zn removal over 80 %). Both materials were very fast-acting (<5 minutes) in achieving metal attenuation. It is envisaged that these adsorptive materials have the potential to be included into structures (such as sand filter beds) that can achieve tertiary treatment of road runoff waters on the upgraded Kuranda Range Road.

Overall, this research demonstrated that annual wet-dry climate cycles control the concentrations, mobility and bioavailability of traffic-derived metals in roadside corridors in the tropics. Metals accumulate in roadside sediments and soils during the prolonged 'dry season' from April to October, and are mobilised by road

runoff waters over the 'wet season' (November to March). Mobile metals are bioavailable to organisms living adjacent to roads. Consequently, remediation strategies that can reduce the dispersal of these contaminants into natural environments are important in road design and maintenance in the tropics. The use of adsorptive materials such as bentonite in sand filter beds is presented as one such remediation option.

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Glossary

AAC – Advanced Analytical Centre (locations at Cairns and Townsville)

ADD – Antecedent-dry-day (Han et al. 2005)

ADT – Average daily traffic

AHD – Australian Height datum

ANSTO – Australian Nuclear Science and Technology Organisation

ALS – Australian Laboratory Services Pty Ltd

AMG – Australian Map Grid

Background levels – The range in values representing the normal concentration of a given element in a material under investigation, such as rock, soil, plants and water (Gregorich et al. 2002).

bgs – Below ground surface

Biological availability (bioavailability) – The readiness of a chemical compound or element to be taken up by a living organism (Gregorich et al. 2002).

BOM – Bureau of Meteorology

CDU – Charles Darwin University

CEC – Cation exchange capacity

Colloid – Particles smaller than approximately 1 μm (White 1997).

Complex/ coordination compound – Complexes or coordination compounds generally consist of one or more central atoms or central ions, usually metals, with a number of ions or molecules, called ligands, surrounding them and attached to them (Snoeyink and Jenkins

1980). Complexes may be nonionic, cationic or anionic depending on charges of central ions and ligands (Snoeyink and Jenkins 1980).

Contamination – Refers to circumstances where a substance is present in the environment, but not causing any obvious harm (Alloway and Ayres 1997).

C_{org} – Organic carbon

CSIRO – Commonwealth Scientific and Industrial Research Organisation

DTPA – Diethylenetriaminepentaacetic acid

Dry season – The months between and including April and October in north Queensland.

EDS – Energy dispersive spectrometry

EDTA – Ethylenediaminetetraacetic acid

First flush – The delivery of a disproportionately large load of constituents during the early stages of the runoff hydrograph (Schueler 1987).

Flocculation – The joining of particles through electrostatic forces or van der Waals' forces (White 1997).

FNQ – Far North Queensland

GBR – Great Barrier Reef

Heavy metals - Heavy metals are those metals, including transition and non-transition metals, possessing densities greater than 6g/cm³ (Alloway 1995). Relative to the alkali and alkaline metals, heavy metals display high ionisation potentials owing to their strong nuclear charge and, as such, are prone to covalent bonding which leads to the formation of stable complexes and compounds (Parker and Rae 1998). They are commonly classed as potentially toxic to life forms, even in very low concentrations (Siegel 2002). However, Hodson (2004) discourages use of the phrase 'heavy metals' and states that it is a "poor scientific term". This is due to many contradictory definitions for heavy metals, based on density, atomic weight and atomic number (Hodson 2004). Despite the controversy

associated with the term, 'heavy metal' is used in this project to refer to the metals Cd, Cu, Pb, Ni and Zn.

Humic substances – Naturally occurring complexing agents that are degradation-resistant secondary organic materials (Manahan 1993; Alloway 1995).

ICP-AAS – Inductively coupled plasma atomic absorption spectrometry

ICP-AES – Inductively coupled plasma atomic emission spectrometry

ICP-MS – Inductively coupled plasma mass spectrometry

Mobility – The ability of particles and substances to move, either by random motion or under the influence of fields or forces (Queensland Department of Environment 1998).

Organometallic compounds – Compounds in which the organic portion of the anion is bonded to the metal by a carbon-metal bond (Manahan 1993).

Platinum-group elements (PGEs) – Platinum-group elements include Ir, Os, Pd, Pt, Ru and Rh (Cabri 1981). Like the heavy metals, the PGEs display relatively high ionisation potentials relative to the alkali and alkaline earth metals. The PGEs investigated in this project are Pd and Pt.

Pollution – Pollution describes circumstances where toxic effects of contaminants have been observed (Alloway 1995).

Risk – The probability that an adverse effect will occur in a person, group or ecosystem that is exposed to a particular concentration of a hazardous agent (*HRAMCS* 1996) (The Health Risk Assessment and Management of Contaminated Sites).

Roadside corridors/environments – Environments immediately adjacent to roads.

RPD – Relative percent difference

rpm – Revolutions per minute

SEM – Scanning electron microscopy

S_{sulfate} – Sulfate sulfur

S_{sulfide} – Sulfide sulfur

TEA – Triethanolamine

TIMS – Thermal ionisation mass spectrometry

TOC – Total organic carbon

Topsoils – The 0-10cm soil layer below the Earth's surface. Commonly includes the O, A and B soil horizons which host a mixture of primary and secondary minerals as well as decaying organic matter and degradation-resistant humic compounds (Alloway 1995; Siegel 2002).

Toxicity – The quality or degree of being poisonous or harmful to plant, animal or human life (Queensland Department of Environment 1998).

Treatment train – A number of measures used in sequence to remediate storm and road runoff waters (Melbourne Water 2005).

Wet season – The months between and including November and March in north Queensland.

Wt % – Weight percent

WTWHA – The Wet Tropics World Heritage Area, located adjacent to the North Queensland coastline.

XRD – X-ray diffraction

XRF – X-ray fluorescence