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## Lead and Cadmium Cycling in a Small Forest Catchment in Japan

By

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K e y w o r d s : Trace metal, pool, flux, Katsura headwater catchment.

#### Summary

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Lead and cadmium concentrations, fluxes, and pools were measured in the bulk precipitation, stream water, vegetation, litterfall, forest floor and mineral soil in a small forest catchment in Japan. At Katsura headwater catchment, runoff outputs of lead and cadmium were much less than precipitation inputs. The input-output budgets indicated that some atmospheric input of lead and cadmium were retained within the catchment. Moreover about 70 % of the lead and cadmium which were taken by trees were reduced again as litterfall to the forest floor. At the present time, forest ecosystems act as sink for atmospheric deposited lead and cadmium in Japan.

#### Introduction

Lead and cadmium have toxic effects at very low levels on plant, animal, and human health. These metals are emitted by anthropogenic activities such as mining, fossil fuel combustion, industrial processes, and waste incineration. Japan is one of the largest users of metals in the world, taking first place in cadmium use and third place in lead. Additionally, trace metals emissions including lead and cadmium are increasing in Asia. Once emitted, trace metals can be transported long distances in air masses and contaminate terrestrial ecosystems (PACYNA & PACYNA 2001).

Sixty-eight percent of Japan is covered with forests. However, there is little information about these trace metals in forests in Japan. Therefore, it is necessary

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to compile information in order to assess the impact of these pollutants on forest environments. The aim of this study was to determine lead and cadmium pools and fluxes in a small forest catchment in Japan.

#### Material and Methods

#### Study area

The Katsura headwater catchment (KHC) (N36°32', E140°18'), Ibaraki, is located in a hilly area about 120 km from the center of Tokyo. The study area spans 210 to 270 m elevation. The mean annual temperature is 13.0 °C and the mean annual precipitation is 1500 mm. The catchment has an area of 2.3 ha. A 38-year-old stand of Japanese cedar (*Cryptomeria japonica*) covers the middle and lower slopes. The upper slope is covered with a deciduous broad-leaved forest comprising three species: *Quercus serrata*, *Prunus jamasakura*, and *Carpinus laxiflora*. The soils are brown forest soils covered with volcanic ash 1 to 2 m thick.

#### Sampling

Bulk (i.e. wet + gravitational dry) precipitation was collected monthly in an open area adjacent to the study area. Streamwater was collected monthly in the study area. Runoff was recorded at a gauging station fitted with a V-notch weir and a continuous water-stage recorder. The water samples were passed through 0.45- $\mu$ m membrane filter. All water samples were acidified with concentrated nitric acid (1:100 v/v) before analysis to promote desorption of metals from the walls of the sample storage bottles.

Mineral soil samples of each horizon were taken from five soil pits excavated in this study area. In the laboratory, the soil samples were dried at room temperature and were sieved through a 2-mm sieve.

Samples of stem wood, stem bark, branches, twigs, foliage, and roots were taken from the four major tree species. Litterfall was collected monthly in five plots in funnel-shaped traps measuring 1 m across and standing 1.5 m above ground level. Forest floor samples (i.e. the organic horizon overlying the mineral soil,  $A_0$ ) were collected within 1.0-m<sup>2</sup> sub-plots. In the laboratory, all vegetation samples were oven-dried in paper bags. Estimates of aboveground and belowground forest biomass were based on allometric relationships between various weight components of individual trees and their diameters at breast height.

#### Digestion and chemical analyses

Soil and plant samples were ground in an agate mortar and then approximately 0.4 g of plant or 0.5 g of soil samples were microwave digested with  $HCl-HNO_3$ .

Lead and cadmium concentrations in the water and soil samples were determined by inductively coupled plasma mass spectrometry, and in the plants samples by inductively coupled plasma atomic emission spectrometry.

#### Results

Figs. 1 and 2 summarize the pools and fluxes of lead and cadmium in the forest catchment (KHC).

Volume-weighted mean concentrations in bulk precipitation were 1169 ng Pb  $L^{-1}$  and 82 ng Cd  $L^{-1}$ . There was no seasonal periodicity of either metal in bulk precipitation. Volume-weighted mean concentrations in streamwater were 26 ng Pb  $L^{-1}$  and 17 ng Cd  $L^{-1}$ . The concentrations were always much lower in streamwater than in precipitation. The annual lead input into this catchment is estimated to be

approximately 23 g ha<sup>-1</sup>y<sup>-1</sup> and estimated lead output is approximately 0.8 g ha<sup>-1</sup>y<sup>-1</sup>. The annual cadmium input is estimated to be approximately 1.5 g ha<sup>-1</sup>y<sup>-1</sup> and estimated cadmium output is approximately 0.2 g ha<sup>-1</sup>y<sup>-1</sup>.

At KHC, the lead concentration in the topsoil ranged between 22.1 and 40.3 mg kg<sup>-1</sup>. The cadmium concentration in the topsoil ranged between 0.19 and 0.46 mg kg<sup>-1</sup>. Both metals were more concentrated in the surface soil than in the deeper soil. Standing pools of lead and cadmium were estimated for a 1-m-deep profile; their values were 99.5 kg Pb ha<sup>-1</sup> and 1.0 kg Cd ha<sup>-1</sup> in Japanese cedar stand and 124 kg Pb ha<sup>-1</sup> and 1.3 kg Cd ha<sup>-1</sup> in broadleaved stand.

Concentrations of lead and cadmium by tree parts followed the pattern fine roots > bark > leaves, twigs > wood > large roots in all species. Total contents in the aboveground vegetation were 191 g Pb ha<sup>-1</sup> and 20.2 g Cd ha<sup>-1</sup> in Japanese cedar stand, and 70.7 g Pb ha<sup>-1</sup> and 5.9 g Cd ha<sup>-1</sup> in broadleaved stand. Total contents in belowground vegetation were 29.5 g Pb ha<sup>-1</sup> and 7.2 g Cd ha<sup>-1</sup> in Japanese cedar stand, and 22.2 g Pb ha<sup>-1</sup> and 9.1 g Cd ha<sup>-1</sup> in the broadleaved stand.

The annual amount of litterfall of Japanese cedar was about twice that of the broadleaved trees. The annual lead fluxes through litterfall were 28.9 g ha<sup>-1</sup> in Japanese cedar stand and 7.2 g ha<sup>-1</sup> in broadleaved stand, and the cadmium fluxes were same in both stands.

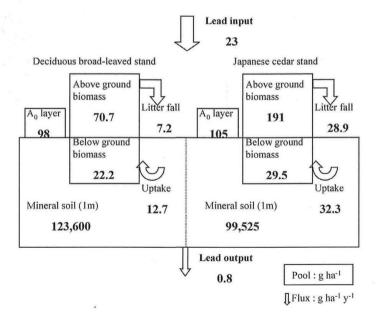


Fig. 1. Pools and fluxes of lead in Katsura headwater catchment (KHC).

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On the forest floor (in the  $A_0$  layer), the metal contents were estimated as 105 g Pb ha<sup>-1</sup> and 5 g Cd ha<sup>-1</sup> in Japanese cedar stand, and 98 g Pb ha<sup>-1</sup> and 10 g Cd ha<sup>-1</sup> in the broadleaved stand.

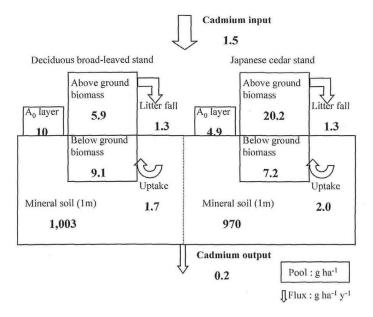


Fig. 2. Pools and fluxes of cadmium in Katsura headwater catchment (KHC).

#### Discussion

Input–output budgets showed that most of the atmospheric inputs of lead and cadmium were retained somewhere within the catchment. Lead retention was very high (>96%) and cadmium retention was also high (86%). These values are similar to those reported for temperate and boreal forest ecosystems (SMITH & SICCAMA 1981, UKONMAANAHO & al. 2001). These results indicate that atmospherically lead and cadmium have accumulated highly within Japanese forest ecosystems.

The mineral soil, forest floor, and plants were the major pools of atmospheric deposited lead and cadmium in the forest ecosystems. Despite the similar lead and cadmium concentrations in plants, these amounts in above ground biomass were larger in Japanese cedar stand than those of broadleaved stand. The reason for this is that the volume of Japanese cedar stand was much more than that of broadleaved stand due to the efficient afforestation.

The highest concentrations were found in the organic surface soil and mineral soil, which are the largest pools for both elements in this catchment. The  $A_0$  layer in forest ecosystem has often been found to be an effective sink for heavy metals, particularly lead (TURNER & al. 1985, BERGKVIST 1987). The amounts of the  $A_0$  layer in the cool-temperate forests are more than that of this study area (TSUTSUMI 1991). In Japanese forest ecosystems, the residence times of these elements in the forest floor are also supposed to be shorter. Therefore, in most Japanese forests, the contribution of the  $A_0$  horizon to the retention of lead and cadmium is relatively lower than in European and North American forests. These results indicate that the upper mineral soil and the circulation of elements through plants and microorganism are more important in the retention of lead and cadmium in Japan. Cadmium, however, is more mobile than lead because it is not as strongly adsorbed by organic matter. Cadmium is moved by cation exchange and is therefore relatively easily taken up by tree roots or lost to leaching (BERGKVIST & al. 1989). Although the transport of cadmium and lead through vegetation is rapid, the slow response of soil and the potential for recycling these trace elements lead to low output from forest catchments.

Lead and cadmium from the atmosphere are retained in the forest ecosystem, which is a sink for these elements. Lead was accumulated and homogenized in the organic-rich fraction of the soil, where it had a residence time of 150-500 years (FRIEDLAND & al. 1992). There is a possibility, however, that accumulated lead and cadmium might be released from forest catchments more rapidly than we expected by global warming or by changes in soil acidity and land-uses, and that forest catchments will become a source of the these metals. Therefore, long-term monitoring of trace metals and further studies in forest ecosystems are needed to obtain more accurate values.

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