Origin of lead in rainwaters from intertropical Africa

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The chemical composition of rainwater is a good indicator of gases and aerosols present in the atmosphere, and also of their sources on a local and regional scale. In order to quantify and identify the different sources of lead in intertropical Africa, rainwaters were sampled over a two year period in four countries with various vegetation zones (Niger: tall and desert grass savannah, Ivory Coast: high grass savannah and rain forest, Cameroon and the Central African Republic: rain forest). Dissolved phase concentrations were obtained after a 0.22 μm filtration. Total phase concentrations (dissolved fraction and particles) were obtained after evaporation and HF:HNO3 digestion. Due to its high sensitivity, accuracy and speed, ICP-MS (Inductively Coupled Plasma-Mass Spectrometer) was used to determine Pb and Al concentrations.

Pb concentrations

First, Pb concentrations in the dissolved and total phase of precipitations from intertropical Africa are compared with precipitation, snow and ancient ice data from other regions (Table 1). In Africa, Pb concentrations in the dissolved phase (0.059–0.25 μg l^{-1}) are: (i) 1.5 to 10 times lower than the concentration found in precipitation from temperate regions of continental sites (Norway, Russia, Finland, Switzerland, France), (ii) equivalent to the concentration found in precipitations from the North Atlantic region (0.094 μg l^{-1}), (iii) 2 to 10 times higher than in Greenland snow, (iv) 15 to 60 times greater than in ancient ice (corresponding to precipitation between 470 and 7760 years ago). As for the dissolved phase, Pb concentrations in the total phase of precipitations from France (3.8 and 14 μg l^{-1}) are 3 to 10 times higher than the level at the Kollo station which shows the highest Pb concentration (1.5 μg l^{-1}). Pb concentration obtained in precipitations from the North Atlantic region (0.20 μg l^{-1}) is quite similar to the results at Lamto and Nsimi (0.26 μg l^{-1}).

Enrichment factors

Mean enrichment factors, calculated with respect to upper crust mean composition (using Al as reference element) are presented in Table 1. Considering a limit value of 10, as indicative of another source in addition to the crustal one, Pb is enriched at Bangui (Ef = 10.5) and at Nsimi (Ef = 12.9) but not at Kollo (Ef = 2.6) and Lamto (Ef = 4.4). However, some Pb enrichment factors calculated for each rain event show values higher than 10 for all the African stations (at low Al concentrations). Pb mean enrichment factors obtained at Bangui or Nsimi (around 10) are lower than in rainwaters from a rural site in France (28.2) and much lower than in precipitations from Paris (354). The Pb mean enrichment factor (11.0) from remote areas (North Atlantic) is of the same order as the ones obtained at Bangui and Nsimi (respectively 10.5 and 12.9).

Pb percentages in the dissolved phase

The percentages of Pb in the dissolved phase (Table 1), obtained with the volume weighted mean concentrations, are variable between the four stations: from 7% at Kollo, which is characterized by a high pH value (5.72) and a high Al concentrations (2312 μg l^{-1}) to 44% at Bangui where the pH and the Al concentration are lower (repectively 4.71 and 219 μg l^{-1}). Pb percentages in the dissolved phase were plotted as a function of Pb enrichment factors and two types of particles were identified: crustal particles characterized by low solubilities (<5%) and enrichment factors close to 1, and very solubles particles (>70%), strongly enriched compared to the silicate upper crust (Ef >100). The samples which show enrichment factors higher than 1 and Pb percentages with a high variance in the dissolved phase (5-80%) contain the two types of particles.

Measurements of Pb concentrations both in the dissolved and the total phase of rainwaters are

TABLE 1. Comparison between Pb concentrations, enrichment factors and phase distributions obtained for rainwaters from intertropical Africa and for precipitation, snow and ice from various sites in the world

	Site	Dissolved phase		Total phase		Ef (Pb)	Dissolved fraction	
		Pb ($\mu g.l^{-1}$)	pH	Al ($\mu g.l^{-1}$)	Pb ($\mu g.l^{-1}$)		Pb	Al
Precipitations								
	Intertropical Africa:							
	Kollo	**0.11**	**5.72**	**2312**	**1.5**	**2.6**	**7.3%**	**0.9%**
	Lamto	**0.059**	**5.09**	**235**	**0.26**	**4.4**	**23%**	**3.4%**
	Bangui	**0.25**	**4.71**	**219**	**0.57**	**10.5**	**44%**	**3.8%**
	Nsimi	**0.094**	**4.88**	**81**	**0.26**	**12.9**	**36%**	**8.4%**
	Norway[1]	2.4	4.6					
	Russia[1]	0.61 - 6.3	4.0 - 5.0					
	Finland[1]	0.56 - 0.84	4.7 - 4.8					
	North Atlantic[2]	0.094	5.20	73	0.20	11.0	45%	13%
	Camargue (France)[3]	2.2		542	3.8	28.2	52%	19%
	Paris (France)[4]	3.3		159	14	354	24%	
Snow	*Greenland*[5]	0.028						
Ancient ice	*Greenland*[6]	0.001 - 0.004				0.8 - 4		

1: Median concentrations from Reimann *et al.* (1997); 2: Volume weighted mean concentrations from Lim and Jickells (1990); 3: Mean concentration from Guieu *et al.* (1991); 4: Volume weighted mean concentrations from Roy (1996); 5: Average yearly concentration from Wolf and Peel (1988) and 6: Ice core concentrations (between 130 and 1290 m) from Hong *et al.* (1994). Pb concentrations in the dissolved phase were obtained as follow: 1 and 3: 0.45 μm filtration; 2 and 4: 0.2 μm filtration and 5 and 6: direct injection by GFAAS (Graphite Furnace Atomic Absorption Spectrometry)

unusual. Pb percentages obtained in the dissolved phase of precipitations from Paris (24%), from the Atlantic Ocean (45%) and from the Camargue region in France (52%) are in the same range as ours results (7.3–44%).

All the results obtained in this study indicate that terrigeneous dust is not the only source of lead in intertropical Africa. An anthropogenic origin for this element (emissions from cars or industries) is clearly identified at Bangui. This origin is characterized by high Pb concentrations in the dissolved phase (>1μg l^{-1}), poor correlations between Pb and the others elements in the dissolved and total phase, Pb individual enrichment factors which can reach a value of 600 and a Pb mean enrichment factor higher than 10 (10.5). Although Pb mean enrichment factors are much lower than 10 for Kollo and Lamto (respectively 2.6 and 4.4), individual Pb enrichment factors are close to 100 and the small correlations including Pb in the dissolved phase suggests a source of lead derived from human activities. The good correlation, in the dissolved phase of Nsimi rainwaters, between Pb and others species like Rb (R = 0.84) or NH4 (R = 0.82), which are well known to be emitted by the vegetation, may indicate a direct biogenic source for Pb or some remobilization processes of anthropogenic Pb deposited onto the vegetation. However Pb concentrations in African rainwaters, are lower than precipitations from industrialized countries. This indicates that the atmosphere is slightly polluted by human activities in intertropical Africa.

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