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Review of pollutant lead decline in urban air and human blood: A case study from northwestern Europe

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ABSTRACT

A review of the transient decline of pollutant lead in the air (Pb_A) and the blood (Pb_B) has been conducted in order to assess the relationship between these environmental reservoirs. We have demonstrated that Pb_A decreased 20 to 100 times more than Pb_B for the past 30 years, suggesting another significant intake besides airborne lead to explain lead accumulated in humans. This trend has also been observed in two blood surveys we have completed in 1976–1978 and 2008–2009 in northern France and Belgium. Nowadays, the mean Pb_B (1.5–3.5 μg/dL) remains at least 100 times higher than the estimated non-contaminated Pb_B. Lead isotope imprints in blood could help decipher specific contamination cases, and were coherent with the decline of Pb_A, but could not help discriminate the source of blood lead owing to the lack of source imprints, especially from dietary intakes. Correlations between recent Pb_B, isotopic imprints and the age of the subjects suggested that lead released from bones has become a significant source of lead in blood. The significant cause for human exposure to lead may have shifted from direct pollutant lead input accumulated in exogenous reservoirs (air and diet) to endogenous lead release from bone tissues consequential to metabolic calcium homeostasis and bone turnover.

1. Introduction

Pollutant lead has largely pervaded urban and forested areas owing mostly to its dispersal into the atmosphere from the use of leaded gasoline since the mid-1920s (e.g., Dorr et al., 1990; Friedland et al., 1992; Janssens and Dams, 1975; Négrel et al., 2015; Nriagu, 1990; Nriagu and Pacyna, 1988; Stille et al., 2011, 2012; Von Storch et al., 2003), resulting in a contamination of the whole population (NRC, 1993; Patterson, 1980; Patterson et al., 1991; Smith and Flegal, 1995). This environmental pollution has been monitored in blood where lead concentrations (Pb_B) have raised until 100 to 1000 times more than the natural uncontaminated levels (Flegal and Smith, 1992; Patterson, 1980; Patterson et al., 1991). Given the short residence time of lead in the blood (less than a month, e.g., Gulson et al., 1994; Rabinowitz et al., 1973, 1976), one would expect Pb_B to closely mimic lead intake. The phasing out of leaded gasoline (1970s and 1980s in the US and western Europe, respectively) has been closely matched by a

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decrease in Pb_B (Annest et al., 1983; Bono et al., 1995; Brody et al., 1994; Ducoffre et al., 1990; Manton, 1985; Schwartz and Pitcher, 1989; Strömberg et al., 1995, 2008, 2014; Thomas et al., 1999; Wietlisbach et al., 1995; Wang et al., 1997), suggesting airborne lead to be a significant source for lead intake. However, this atmospheric contribution was found highly variable in pioneering studies from the 1970s ($7 \pm 3\%$ to $41 \pm 3\%$) (Colombo et al., 1988; Manton, 1977) when gasoline lead content reached its climax (Nriagu, 1990). Because lead in the blood and the atmosphere have similar residence times, one would expect both reservoirs to mimic the same patterns with time, unless there is another significant lead intake in humans. The extent to which changes in Pb_B could be related to the fall of lead content in the air could also be assessed using stable lead isotopes and their transient imprints.

Lead has four stable isotopes (masses = 204, 206, 207, and 208), the last three being end-members of the radiogenic U–Th decay chains (Doe, 1970). Because of variations in the initial U–Th content of mineral reservoirs, and the different decay rates of U–Th parent isotopes, crustal material and lead ores display specific lead isotopic ratios. This property has allowed to distinguishing between natural and pollutant Pb sources as well as various industrial and automotive lead sources in blood (Chaudhary-Webb et al., 1998; Glorennec et al., 2010; Gulson, 2008; Gulson et al., 2006; Keinonen, 1992; Kurkjian and Flegal, 2003; Manton, 1973, 1977; Rabinowitz, 1987; Rabinowitz and Wetherill, 1972; Soto-Jimenez and Flegal, 2011; Tsuji et al., 2008; Yaffe et al., 1983). While thousands of Pb_B values have been determined within the past 40 years, only a few hundreds of lead isotope imprints have been measured from less than 30 published articles. The relative scarceness of lead isotope investigations in the blood as compared to Pb_B is attributable to analytical difficulties (i.e. the need for chromatographic extraction in dedicated clean laboratories and the use of mass spectrometers, equipped with multi-collection faraday-cups and thermal-ionized or plasma emissions). A review of investigations concerning the use of stable lead isotopes in humans can be found in Gulson (2008).

After the phasing out of leaded gasoline, Gulson et al. (1996) suggested that Pb_B isotopic imprints for low Pb_B levels ($< 5\ \mu g/dL$) could be altered by environmental exposures that were previously hampered by the overwhelming petrol lead intake. More recently, Gulson et al. (2006) suggested that lead from diet intake might have become the main source of Pb_B. Manton et al. (2005) came to the same conclusion but could not distinguish between household dust and diet sources to explain Pb_B. Nowadays, while lead in diet is generally of no threat anymore (Bolger et al., 1996; Egan et al., 2002; Manton et al., 2005), some specific diets could constitute significant lead sources (Manton et al., 2005; Ndung'u et al., 2011; Ranking et al., 2005; Rosman et al., 1998; Scelfo and Flegal, 2000). Partial bone mineral resorption may also favour the release of lead from bone tissues to blood depending on gender, age and environmental constraints owing to normal calcium homeostasis, pregnancy, osteoporosis, and could contribute to 10–70% of Pb_B (e.g., Chamberlain et al., 1978; Gulson, 2008; Gulson and Gillings, 1997; Gulson et al., 1995, 1996; Hernandez-Avila et al., 1998; Keller and Doherty, 1980; Manton, 1985; Manton et al., 2003; Rabinowitz, 1991; Rabinowitz et al., 1973, 1974, 1976; Silbergeld and Watson, 1993; Silbergeld et al., 1988, 1993; Smith et al., 1996; Thompson et al., 1985).

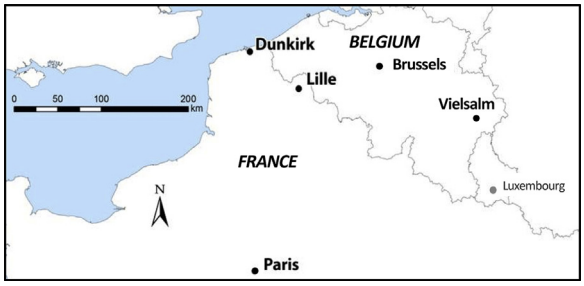

Fig. 1. (Color online.) Cities (in bold) where Pb_A were reviewed and Pb_B were measured.

The significant decline of airborne lead sources has raised specific questions regarding:

(i) the contribution of atmospheric lead (Pb_A) to Pb_B in urban areas;
(ii) the declining trends of airborne lead and Pb_B in countries where the phasing out of leaded gasoline was initiated at least 15 to 20 years ago;
(iii) the relative increase of exogenous non-atmospheric and endogenous lead contribution to Pb_B.

In order to investigate these issues and the relationship between Pb_A and Pb_B levels, we have reviewed mean annual Pb_A values and the corresponding Pb_B ones for the past 30 to 40 years in countries where tetraethyl lead has been banned from gasoline for at least 15 to 20 years. To supplement this database, we measured Pb_B and isotopic imprints in 1976–1978 and 2008–2009 in northwestern Europe (Brussels, Vielsalm and Dunkirk, Fig. 1) where urban monitoring stations could provide mean annual Pb_A since the late 1970s. The area of concern (in France and Belgium) is among the region that exhibits the highest atmospheric particulate matter (PM10 and PM2.5) concentrations in Europe (CAFE, 2005; EMEP, 2011). Thus, for people living in these regions, and especially in urban areas, the ambient air potentially could remain a possible pathway for Pb_B. The review of combined Pb_B and isotopic imprints over decades during which lead was banned from gasoline might provide insights into major changes in human lead impregnation.

2. Material and methods

2.1. Atmospheric and blood reviews

Annual mean Pb_A values were compiled (Fig. 2) from Lille (ATMO Nord-Pas-de-Calais network), the closest available monitoring station to Dunkirk where blood samples were taken as part of this study, and from Brussels (Bruxelles Environnement–IBGE network). Because mean Pb_A values were not available prior to the early 1980s in Lille, monitoring stations from Paris (AIRPARIF