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Many of us are familiar with the unwelcome sight out of the aeroplane window: a brownish haze looming over a major city as you glide in towards the airport. Many components of this air pollution are not benign and understanding where they come from, how they affect us, and how to reduce them are important health issues. While urban centres are major contributors to this pollution problem, the aerosols are dispersed much more

undertake detailed aerosol characterisation and to develop predictive models for PM_{10} concentrations and their sources. Together with colleagues at Kings College London who are engaged in PM_{10} measurement and modelling, the NERC Isotope Geoscience Laboratory at the BGS is studying the lead isotopes of PM_{10} particles from urban aerosols collected in London over a four-year period straddling the time that leaded

Airborne pollution

The evolving lead isotope signature of PM_{10} atmospheric aerosols

by Steve Noble¹, Matt Horstwood¹ and Steve Smith²

widely, affecting even remote areas far from busy city life.

Many of the pollutants are gaseous but there are also significant amounts of suspended particles, or aerosols. Urban aerosols are a mixture of relatively insoluble inorganic and organic material and highly soluble species such as nitrate, sulphate and ammonia. They have diverse sources including suspended soil particles, vehicle emissions, power plants, incinerators or factories, and it is obvious that these are often locally derived. What is perhaps less widely appreciated is that there is good evidence that the aerosols can also travel huge distances, for example across the Atlantic Ocean or Europe, catching a free ride with prevailing atmospheric winds.

Aerosol particles are small enough to be inhaled and deposited in the lung. Particles with diameters less than 10 micrometres (PM_{10}) can lodge deep into the lung and have been associated with deleterious effects on human health. The UK National Air Quality Strategy has been designed to seek mechanisms to significantly lower the ambient PM_{10} concentrations by 2010.

To understand the origin and behavior of these aerosols it is important to

petrol was phased out. This work is, in part, a pilot study to work out analytical protocols but it is also establishing a baseline lead isotope signature for this metropolis at a time when significant shifts in isotope composition may be occurring. In general, it is known that lead is a toxic pollutant and general tracer for anthropogenic contributions to aerosols. London is a particularly interesting place to address this issue because previous studies revealed local and distal PM_{10} sources. Also the PM_{10} particles related to traffic emissions have a strong association with lead, antimony, zinc, copper and iron, making lead isotopes a sensitive aerosol provenance indicator for this particular source.

There is considerable natural variation in the isotopic ratios of lead, arising from the radioactive decay of uranium and thorium. Ore deposits formed at different times during Earth history thus have ‘frozen in’ contrasting isotopic signatures. The mining of ores from distinct geological terranes leads to variations of lead isotopic compositions used in industrial processes, and in turn to that of vehicle exhaust, depending on the source of lead additives to fuels. Significantly for pollution studies, the landmark works by Bollhöfer and



colleagues at Curtin University of Technology in Perth show that, on a global scale, there is a broad provinciality to anthropogenic lead isotopes roughly bounded by end-member isotope compositions of Australian, western Canadian, US, Mexican and east Asian lead ore deposits. At a given time, an area such as eastern Europe or the UK can have marked lead isotope differences in atmospheric aerosols. It has been observed that the compositional profiles of regions are not static, but within a short enough timescale they can be used to infer where the lead, and by inference the aerosols in general, are coming from.

Our investigations of London aerosol sources started with PM₁₀ obtained during December 1998 and December 1999 from a busy urban roadside location in Marylebone Road, central London. PM₁₀ samples were collected on quartz-fibre filters (*see top right*). The lead isotope compositions of the deposits were analysed by multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) in two ways. The first method was to leach lead out of the filter deposits with acid and analyse the lead-carrying solutions on the MC-ICP-MS. The second method employed laser ablation to ‘dust’ the filter surface, liberating PM₁₀ particles and feeding them directly into the MC-ICP-MS. Both analytical approaches yield similar results.

The data (*bottom right*) indicate how there has been a major impact on lead isotope composition accompanying the gradual removal of leaded fuels. This is seen as a large shift in isotope composition since 1994–95. Similar lead-evolution trends have been observed elsewhere in Europe and highlight the importance of long-term monitoring when assessing regional lead isotope compositions. The current lead isotopic signature of London PM₁₀ emissions resembles that being emitted from north-eastern Europe and Russia during 1994–99. This could be a plausible contributory source of lead-bearing aerosols in London following the reduction of direct UK emissions arising from the elimination of lead additives in fuels.

So what is next? Work continues with 2000 and 2001 London samples to fully characterise the diminishing impact of leaded petrol and elucidate an increasing significance of other local and distal

sources. At the end of the study we will be able to contribute up-to-date baseline information for future global aerosol assessments. And it is hoped that, in turn, our work will assist modellers to more rigorously assess UK contributions to the national PM₁₀ budget and to monitor the nation’s progress in reducing these pollutants. ■



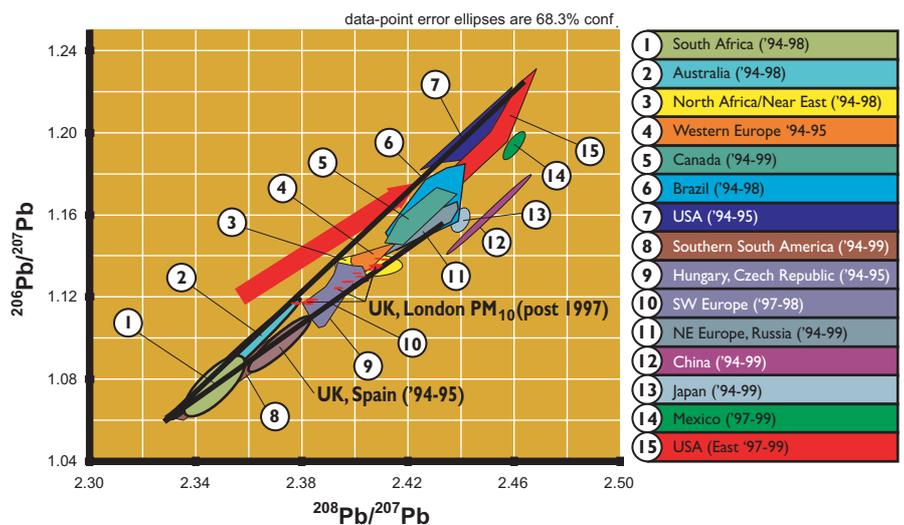
Photomicrograph of quartz-fibre filter, showing the raster sampling pattern produced by laser ablation during ‘dusting’ to remove the sample from the filter substrate. Width of each rastered line is 200 micrometres.

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Global PM₁₀ lead isotope signatures (based on Bollhöfer & Rosman, *Geochimica et Cosmochimica Acta* **65** 1727-1740). London data between 1998 and 1999 are small red ellipses and the arrow indicates the shift in isotopic composition following the phasing out of lead additives to petrol.