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RECONSTRUCTION OF LEAD (Pb) FLUXES IN EUROPE DURING 1955-1995 AND EVALUATION OF GASOLINE LEAD-CONTENT REGULATIONS

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1. INTRODUCTION

The Institute of Hydrophysics of GKSS Research Center is developing the tools and expertise required for both past reconstructions and future predictions of substance fluxes on a regional scale and over a period of several decades, and to evaluate their impacts to society. The goal is to build our capacity to compare between scenarios, be it scenarios of substance-emission regulations, economic development, or climate change. In particular, alternative emission-regulation scenarios may in the future be compared and rated as to their predicted societal impacts.

The *tools* required for flux reconstructions or predictions are climate simulation models and environmental transport models. Transport models for the atmosphere, land, and freshwater bodies permit estimation of substance concentrations in these environmental compartments. The climate simulation models are needed for forcing of the transport models. The *expertise* required is multidisciplinary. It includes understanding of the physical and chemical processes undergone by the substance of interest, in order to select, calibrate, and apply the transport models. It also includes the ability to evaluate the impacts to society of the estimated substance concentrations. The involvement of both natural scientists and socio-economists is hence required.

As our first study we consider the case of lead (Pb). There are four reasons why lead represents a good prototypical example:

- First, as an approximately conservative particle-bound element, lead serves as a marker of particle transport routes and facilitates our understanding of how far and how fast particles move along these routes. Transport models that are set up and validated for lead may in the future be used for other particle-bound substances, after adjusting for their chemical transformations.
- Second, nearly all lead found in the environment today (e.g. in soils, lake sediments and ice cores) has anthropogenic origin, and its bulk is due to the combustion of leaded gasoline by road traffic, mostly after 1955. Reliable estimates for 1955 to present are available for both atmospheric lead emission rates (section 2.1) and climatic conditions in Europe (section 3). Thus, for lead there is the rare opportunity to reconstruct from sources to sinks nearly the entire history of present contamination levels.
- Third, data on lead concentrations in air, soils, sediments and ice cores, as well as estimates of atmospheric lead deposition rates, are available for various European locations (mostly from the 1970's to present), permitting model validation (section 2.3).
- Fourth, the lead content in gasoline has been subjected to a series of increasingly stringent regulations in Europe, and the 1998 Aarhus Treaty has established the prohibition of leaded gasoline sales in all of Europe after the year 2005 (section 2.2). These represent suitable case studies for impact evaluation of past and future regulations. Moreover, the sharp rise in lead emissions up until the mid-1970's and the sudden drop that followed the introduction of each new regulation provide the opportunity to study how fast and how marked a change was seen in environmental lead concentrations.

The "Lead Project" ("Bleiprojekt") uses simulation models of climate and lead transport at the European scale for the 40-year period 1955-1995. The impact study concentrates on the catchment of the river Elbe (ca. 150 000 km^2 with an upstream portion in the Czech Republic and its greater part in Germany). The various project components are detailed in separate sections below.

2. DEVELOPMENT OF INVENTORIES

2.1 Inventory of lead emissions

Expert estimates of European atmospheric lead emissions are available for 1955-1995 at a spherical resolution of 0.5^o (roughly, 50 km). These estimates and the methodology by which they were obtained are summarized in Pacyna and Pacyna (1999). Estimates are provided for the years 1955, 1965, 1975, 1985, 1990 and 1995 (Figure 1). All major lead sources were included in these estimates, namely, road transport, non-ferrous metal manufacturing, stationary fuel combustion, iron and steel production, cement production, waste disposal, and miscelaneous sources. Road transport represented by far the largest source, accounting for about 50% of total European emissions in 1955, 75% from the mid-1970's through the mid-1980's, and 69% in 1995. Therefore, the emission maps in Figure 1 in large part reflect the history of gasoline consumption and lead-content emission regulations, summarized in the next section.

Estimated road transport lead emissions in Europe totalled about 31 thousand tonnes in 1955, nearly quadrupling to 119 thousand tonnes in 1975. As a result of gasoline lead content reductions, road transport emissions have henceforth declined to about 19.5 thousand tonnes in 1995, despite the continued rise in gasoline consumption. The Aarhus treaty (COWI and DTI, 1998) signed in 1998 by nearly all European governments establishes the exclusive use of unleaded gasoline in Europe by the year 2005.

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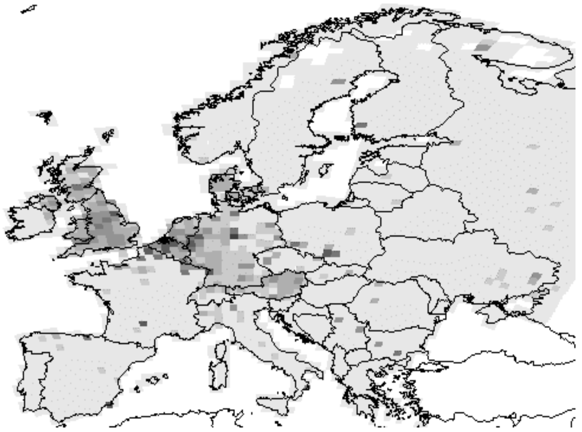
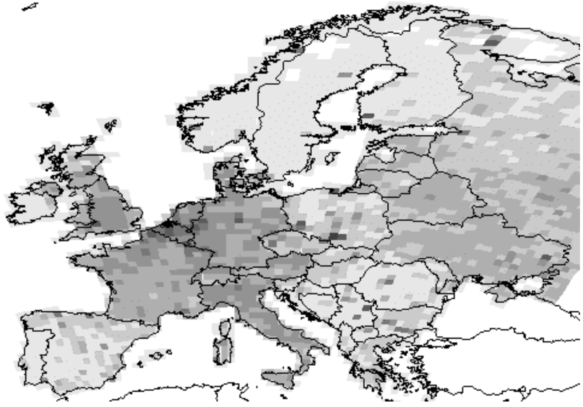
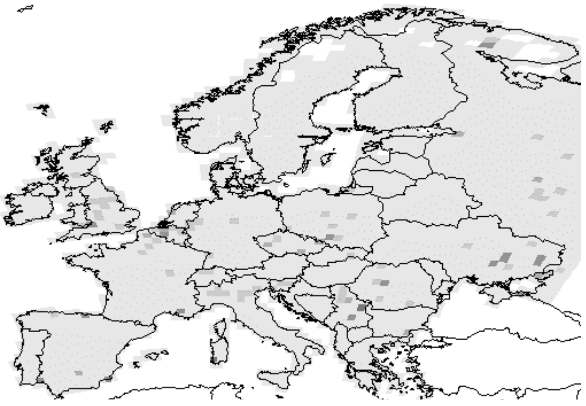


1955 (total: 62 532 tonnes Pb)



1975 (total: 159 233 tonnes Pb)



tonnes **1995** (total: 28 390 tonnes Pb)

	tonnes
░	0 - 25
░	25 - 50
▒	50 - 100
▒	100 - 250
▓	250 - 500
▓	500 - 1000
█	1000 - 1500

Figure 1. Estimated lead emissions in selected years. Regulation of gasoline lead content contributed greatly to bring 1995 emissions to under 1955 levels, after the sharp rise leading to the mid-1970's, and despite continually increasing gasoline sales. (Data source: Pacyna and Pacyna, 1999).

2.2 Inventory and analysis of gasoline-lead legislation

The history of European gasoline lead content regulations and their motivations was reviewed by Hagner (1999). It has two main periods: its rise from the 1930's to the 1970's, and its regulated stepwise reduction from the 1970's to present.

With a gasoline lead content of 0.6 g/l and ever-growing road traffic, automobile lead emissions rised sharply in Europe up to the mid-1970's (section 2.1). Germany, where in the 1970's environmental concerns weighed heavily in national politics (Peters, 1980), was the first to impose gasoline-lead restrictions. Starting in 1972, German production and importation of gasoline with more than 0.4 g Pb/l was prohibited, and starting in 1976 the more strict limit of 0.15 g Pb/l was imposed (Figure 2). The European Union (EU) modestly fixed its limit at 0.4 g Pb/l starting 1978.

In 1983, "unleaded" gasoline (0.013 g Pb/l) was introduced in Germany and its exclusive usage was highly desired because a new combustion catalyst which reduced NO_x, CO and C_xH_y emissions was averse to lead. These gases were mass pollutants thought to pose a threat to forests, and the German government wanted to introduce car-emission regulations as strict as those already in place in the US and Japan (Deutscher Bundestag, 1984). However, it was now not possible to prohibit the sale of leaded gasoline in Germany because the EU precluded trade restrictions among its members. Instead, Germany introduced tax incentives for unleaded gasoline in 1984, and in 1985 its availability at all German gas stations became mandatory. Enhanced tax incentives in 1986 made German unleaded gasoline cheaper than the leaded variety, and its market share in this country has increased steadily thereafter, approaching full share today (about 98%).

In addition to pursuing national policies, Germany also pressed the EU for a European bill. Germany's concerns included transboundary pollution, cross-border road traffic and, finally, the viability of its automobile export industry (which had adopted the

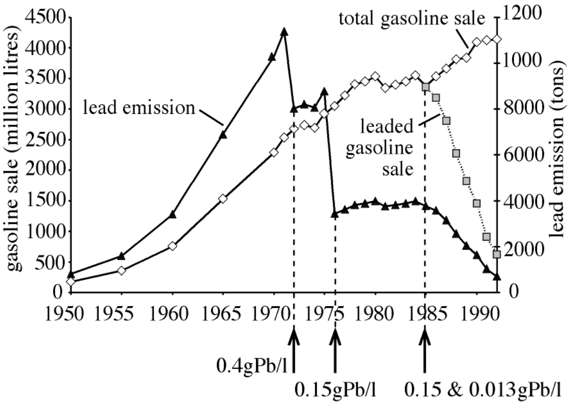


Figure 2. Response of automobile lead emissions in Germany to increasingly stringent gasoline-lead regulations. Despite continually rising total gasoline sales, lead emissions dropped dramatically in 1972 and 1975 following lead content reductions, and have decreased steadily after 1985, as unleaded gasoline concquered the market. (Data source: Mineralölwirtschafts-verband, 1998).

new lead-averse catalysts). In 1985, the EU mandated that by October 1989 Super unleaded gasoline be available for sale in all member states, alongside the leaded variety. Moreover, member states were asked to voluntarily adopt a 0.15 g Pb/l limit. While adherence to unleaded gasoline was quite prompt in Italy and the UK, France offered strong resistance, partly to protect its small-car export industry. In 1987, all member states were allowed to prohibit national production and sales of leaded 92-octane gasoline. Observable damage to public health and the environment was claimed (Rat der Europäischen Gemeinschaften, 1987). We have been unable to find documental evidence of observed damage at the blood lead concentrations reported by the Human-Biomonitoring Commission (1995) (section 5).

Similar arguments led to the signing, by nearly all countries in Europe, of the 1998 Aarhus treaty (COWI and DTI, 1998), which stipulates the exclusive use of unleaded gasoline in Europe by the year 2005.

2.3 Inventory of lead concentrations in the environment

An inventory of available European measurements of lead concentrations in the natural environment and in human blood has been compiled. Contents in the natural environment are needed to evaluate the performance of our simulation models. All lead measurements collected may be used to evaluate the benefits of gasoline-lead policies (section 5).

If our model estimates are in approximate accord with observations, the models will be considered validated. This validation test is non-trivial, not only owing to the simulation period length of 40 years, but especially because of the striking changes in lead emission rates that took place during this period. Can our simulation models accurately predict how much and how fast the different environmental compartments (e.g., air, soils) were affected by the sharp rise in emission rates leading to the mid-1970's? Can they reproduce the corresponding increase in air lead concentrations at various European locations? Do they accurately describe the rates of lead accumulation in soils, in stream sediments and in ice? And can they accurately reproduce the decline in environmental concentrations and accumulation rates resulting from decreasing emission rates after the mid-1970's?

The principal lead data are listed below. "LC" stands for "lead concentration."

- LC in air and rainwater at 32 monitoring stations across Europe (but with weak coverage of some areas): monthly means for 1989-1996 (source: EMEP). Annual means for Denmark for 1979-1987 (source: Danish Dep. of Atmospheric Environment).
- LC in aerosols: monthly means for Germany for 1970-1997 (lead-monitoring network of the German Environment Office); and annual means for Niedersachsen province for 1994-1997 (lead-monitoring network of the State Ecology Office). Annual means at various monitoring stations across Europe for 1993-1994 (source: EMEP).
- LC in organic soils and on tree leaves and needles in the German public forests: samples collected for every 8x8 km^2 unit in Germany, from 1987 to 1993 (source: Institute for Forest Ecology and Statistics).
- LC in the suspended matter of the river Elbe: monthly means at the ARGE monitoring stations for 1984-1998 (source: GKSS).
- LC in sediment cores of the Elbe river bed and floodplain, from locations in Germany and the Czech Republic, the oldest layer dating from 1549 (sources: GKSS and German Federal Office of Hydrology).
- LC in stream waters and stream sediments: samples taken for every 3x3 km^2 unit in Germany from 1977 to 1983 (source: German Federal Office for Geosciences and Raw Materials).
- LC in human blood in Germany in 1979-1997 (source: Schleswig-Holstein State Office for Nature Reserve and Environment).
- LC in mussels from the German Wadden Sea 1985-1997 (German Environmental Sampling Databank).
- LC in plants and soils in Germany (German National Environment Office).

Among data inventoried so far, only the Elbe fluvial sediment cores include dates prior to the 1970's (Figure 3). Concentrations in these cores are markedly affected by industrial effluent discharges. It is air concentration which follows most closely the atmospheric emissions reductions (Figure 4).

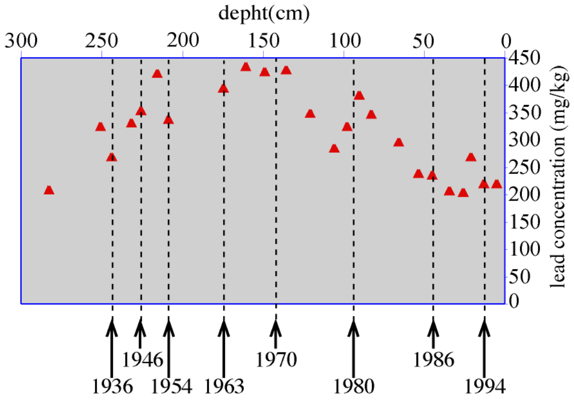


Figure 3. Lead concentration in 20 μm-thick dated layers of an Elbe river sediment core (located in Tangermünde). The high concentrations circa 1950 are largely due to lead-carrying industrial effluent discharges. The increase in the early 1990's may reflect boosted industrial production in the Czech Republic after its shift to a market economy. (Data source: Prange, 1997.)

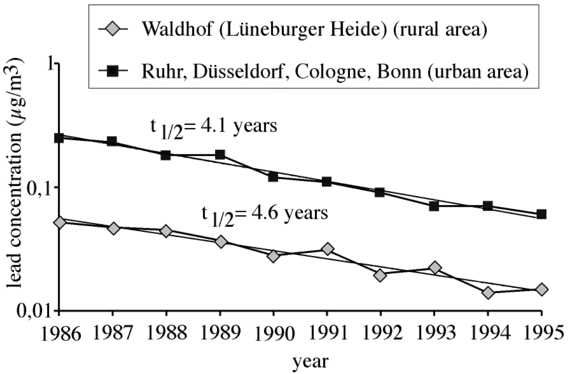


Figure 4. In Germany, air lead concentrations in both urban and rural areas have decreased exponentially since 1986, as the market share of unleaded gasoline increased, requiring only 4-5 years to drop by half. The vertical axis of this plot has logarithmic scale. (Data sources: Umweltbundesamt, 1998, and Landesamt Nordrhein-Westfalen, 1998.)

3. CLIMATE RECONSTRUCTION

The climatic conditions over Europe during 1955-1995 were estimated (or "reconstructed") by the global "reanalyses" of the US "National Centers for Environmental Prediction" (NCEP) (Kalnay et al., 1996). The NCEP reanalyses are in accord with the available point observations, and are sometimes referred to as "observed states." However, not only are their smaller-scale features subject to uncertainty but also, having 2° spherical resolution, they are insufficiently detailed for forcing the atmospheric lead transport and deposition model with accurate representation of the Elbe catchment. To obtain a 0.5°-resolution atmospheric data set we perform a "regionalisation" of the NCEP reanalyses. This is done through dynamical downscaling using the "spectral nudging" technique (von Storch et al., 1999).

Regional-scale climate statistics are conditioned by the interplay between continental-scale atmospheric conditions and such regional features as marginal seas and mountain ranges. The main task in regionalisation is to retain the large-scale features, but to add the regional detail related to physiographic features. The current standard regionalisation procedure is to force a regional climate model to satisfy boundary conditions defined by the reanalyses data. Boundary agreement does however not guarantee large-scale agreement inside the spatial domain, and at times marked deviations from large-scale reanalyses features occur.

Our spectral nudging technique forces the regional climate model to satisfy not only boundary conditions but also the large-scale features inside the domain. In order to force agreement with the large-scale but not the small-scale features, the model must be able to distinguish between the two. For this purpose, the reanalyses data is first subjected to spectral decomposition. A pre-defined critical wavelength λ^* then separates the large- and small-scale spectral domain. There is so far no objective way to define λ^*, but it must be several-fold larger than grid size. The regional climate model is forced to accept (i.e., be "nudged" to) the large-scale reanalyses features. This is achieved by assigning a high value to the nudge coefficient, η, for wave lengths longer than λ^*. For wave lengths shorter than λ^* we set η equal to zero.

We use the regional climate model REMO (Jacob et al., 1995; Jacob and Podzun, 1997), a grid-based model that applies discretized primitive equations in a terrain-following hybrid-coordinates system. The finite differencing scheme is energy preserving. The prognostic variables are surface pressure, horizontal wind components, temperature, specific humidity and cloud water. A soil model is added to account for soil temperature and water content. Horizontal resolution is 0.5° and the domain contains 91 x 81 grid points.

So far, a pilot simulation of 3 months (January-March 1993) has demonstrated the success of the spectral-nudging technique. The large-scale features of the global reanalyses (which are updated every 6 hours) are retained and local detail is added without suppressing short-term variability. Figure 5 demonstrates this success by displaying surface wind components at the oil platform Ekofisk in the central North Sea. In-situ observations and REMO reconstructions are remarkably similar. REMO will be integrated over 40 years and provide detailed representation of the past weather states.

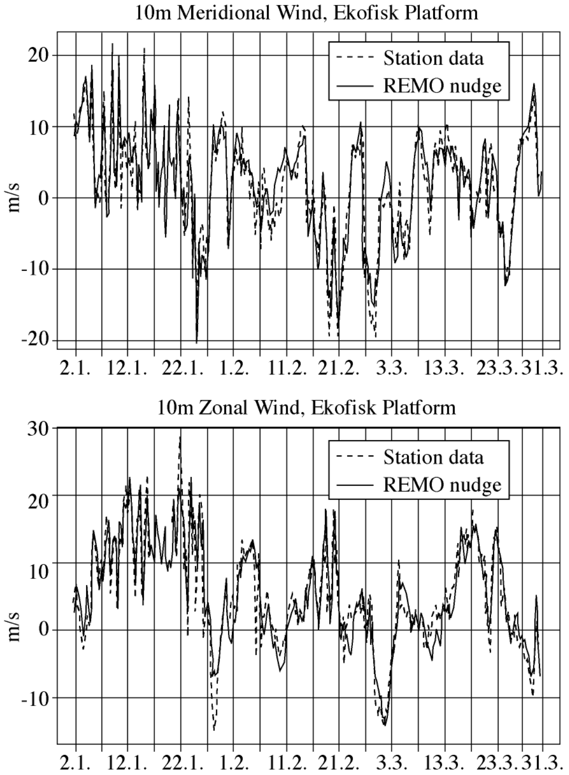


Figure 5. Meridional and zonal wind at Ekofisk (central North Sea), as recorded locally and as simulated.

4. ENVIRONMENTAL TRANSPORT MODELING

Lead attaches to airborne particles and these can be carried over long distances by wind. Long-distance transport is evidenced by the lead contents in the layers of remote ice cores in central Greenland, where the entire history of European lead smeltering is recorded by elevated lead levels (Hong et al., 1995). Confirmed by isotopic analysis to be of anthropogenic origin (Rosman et al., 1995), lead contents of 2-3 pg/g in ice layers dating from 500 BC to 300 AD were generated by ancient Greek and Roman smeltering, 4 pg/g by the Middle Ages and Renaissance, 10 pg/g by the Industrial Revolution, and 50 pg/g in the 19th century. The most part of the 100 pg/g reached in the 1960's (200 times the estimated natural value) and 150 pg/g circa 1984 have been chemically attributed to the organolead additives in gasoline.

Airborne lead particles reach the earth surface by either dry or wet (rainfall) deposition. The atmospheric transport and deposition of lead over Europe has been successfully modelled at 1.5° spherical resolution (roughly, 150 km) using a two-dimensional backward-trajectory (Lagrangian) model (Krüger, 1996). Predicted air lead concentrations and deposition rates compared favorably with observations. However, at 0.5° resolution, backward trajectories are markedly not mass conservative. This is because at this finer scale it often occurs that all back-trajectories miss a lead source, whose emitted mass is then lost by the model.

We use the two-dimensional atmospheric transport and deposition model TUBES (Costa-Cabral, 1999) which uses flow tubes instead of linear trajectories to represent advection from diffuse sources, and guarantees mass conservation at any scale. A flow tube has the capability to widen or narrow when wind directions are divergent or convergent, respectively (Figure 6).

We use TUBES in the forward mode (backward flow tubes can also be used). Forward linear trajectories are used for point sources whose exact location coordinates are specified. Flow tubes are used for diffuse sources, such as road traffic. When the lead emission rate by road traffic is given for a grid box, it is assumed to be uniformly distributed over the grid box's surface. The flow-tube representation corresponds to the limit case where the number of linear trajectories is increased indefinitely, one trajectory for every point in the grid box.

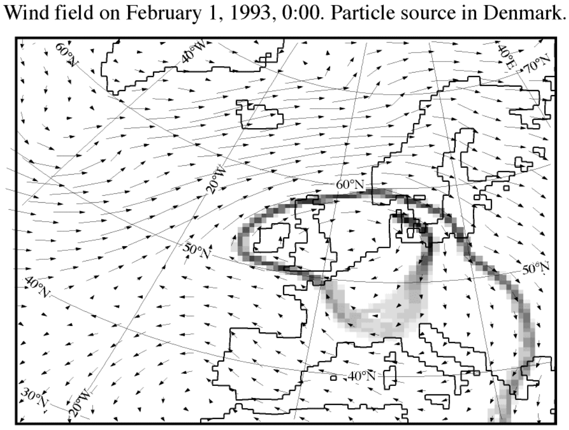

Wind field on February 1, 1993, 0:00. Particle source in Denmark.

Figure 6. Illustration of a flow tube. The wind field is assumed not to change in time and there is no deposition. A single grid box tracer source in Denmark is assumed to emit at a constant rate. The forward flow tube shown first widens as it crosses a region of divergent flow directions, and later narrows as directions converge. Tracer concentrations along the plume (i.e., the flow tube) decrease and increase, correspondingly. The flow tube spans a surface area of about 680 000 km^2. The surface area of all grid boxes transversed by a linear trajectory (not shown) for this same example is only about 370 000 km^2. The linear trajectory affects a smaller area and predicts higher tracer concentrations.

The input data for TUBES is *(1)* the estimated lead atmospheric emission rates (section 2.1), *(2)* two-dimensional wind fields at the 925 hPa level (roughly, 800m altitude) updated every 6 hours, and *(3)* the depth of the atmospheric mixing layer. Inputs 2 and 3 are provided by the climate reconstruction (section 3). The 925 hPa level wind fields are considered to be representative for advection in the atmospheric mixing layer as a whole (Petersen et al., 1989).

Once deposited onto the land surface, lead-carrying particles may be transported overland by runoff and wind. Some will reach a stream channel and be carried by fluvial transport. Others will be deposited on a low-slope terrain area before they can reach a stream channel. Local terrain slope, runoff intensity and particle-size distribution are the key variables determining the rates of lead delivery to the stream network. Smaller particles, having larger specific surface area, have higher lead contents per unit mass (expressed by the sediment "enrichment ratio"). They are also more easily transported by runoff, originating higher sediment "delivery ratios" to channels.

5. SOCIETAL IMPACTS OF GASOLINE-LEAD REGULATIONS

The more direct impacts of gasoline-lead regulations on the German economy were analyzed by Hagner (1999). German mineral oil and automobile markets were most directly affected. The tax incentives granted to unleaded gasoline (section 2.2) benefitted both gasoline traders and consumers, but the new distribution system costs acted to the detriment of medium-sized gasoline traders and independent importers. In the automobile market, a competitive advantage resulted from those producers who already had experience manufacturing

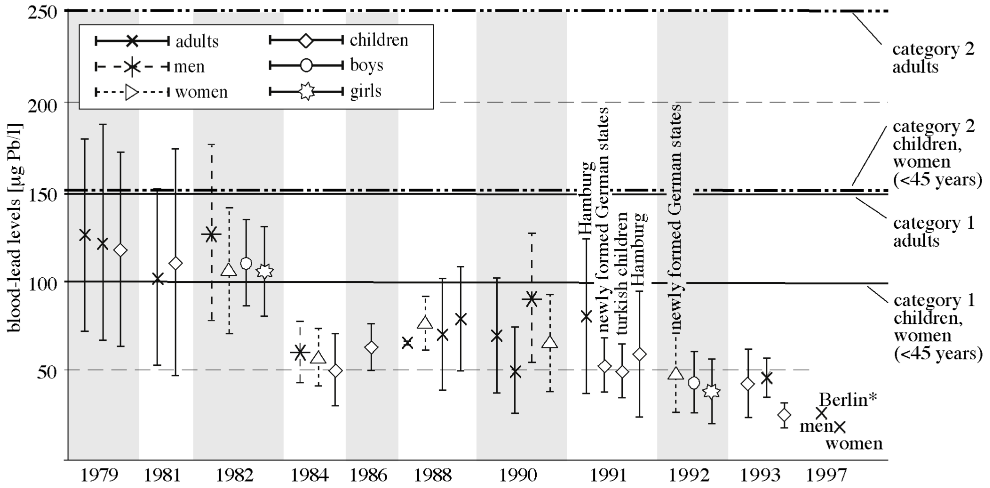


Figure 7. Lead concentrations in human blood in Germany, from various studies. Samples were taken within 2 years of the year shown on the plot. Categories 1 and 2 thresholds were redefined by the German Human-Biomonitoring Commission, most recently in 1995. *Category 1:* normal burden. *Category 2:* no health dangers expected, but controls recommended. For people with blood lead levels above category 2, health dangers cannot be ruled out and controls are recommended (Data source: Heinzow et al., 1998)

automobiles with the new catalysts for the US market.

Lead-content regulations had no measurable effect on economic indicators such as unemployment level, economic growth, price stability and foreign trade balance. Innovation was encouraged and competition was strengthened, so that collaboration within the gasoline and automobile markets was weakened (Hagner, 1999). Costs of gasoline-lead reduction were hardly passed on to fuel consumers, hence neither high-income nor rural populations were burdened.

A significant decline in lead content in human blood was observed in the 1980's and 1990's for all population groups (Figure 7). This decline is due to lowered environmental lead concentrations, including in food items such as fish and molluscs. Categories 1 and 2 in Figure 7 were defined by the German Human Biomonitoring Commission (1995), which determined that values below category 2 are not expected to cause health dangers. According to this Commission, blood lead levels in the general population have not been high enough in the period of study (1979 to present) to cause acute health hazards.

However, no safe threshold has been identified under which the adverse effects of lead cannot be detected. These effects include (from Lovei, 1997): *(1)* in pre-natal exposure, reduced birthweight and skeletal growth, disturbed mental development, spontaneous abortion and premature birth; *(2)* mental and motor affections in children; and *(3)* hypertension and cardiovascular problems in adults.

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