Four Decades of Gasoline Lead Emissions and Control Policies in Europe: A Retrospective Assessment

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Abstract

Over decades, large amounts of the neurotoxin lead was released into the European environment, mostly from gasoline lead additives. Emissions were growing unabatedly until the 1970s, when a series of regulations on the allowed gasoline lead content were adopted. As a result, in the 1990s most gasoline contained only small amounts of lead. We have examined this case of environmental pollution and regulation, and performed a retrospective assessment of the extent of regional-scale lead pollution and the effects of gasoline lead regulations in Europe. With the help of a regional climate model, NCEP re-analyses, spatially disaggregated lead emissions from road traffic and point sources, and various local data, the airborne pathways and depositions of gasoline lead in Europe since 1958 were reconstructed. It turns out that this approach is successful in describing the time-variable, spatially disaggregated deposition of gasoline lead. Additional data from analyses of concentrations in biota, including plant leaves, mussels and human blood, allows an assessment about the impact of the lead phase-out on the quality of the environment. Demonstrating the success of the lead policies, concentrations in leaves and human blood has steadily declined since the early 1980s. At the same time, the economic repercussions that had been feared did not emerge. Instead, the affected mineral oil and car manufacturing industries in Germany (our case-study) were able to deal with the effort without incurring significant extra costs. We suggest that our method of quantitatively reconstructing and anticipating fluxes and depositions of substances can be applied to other relevant substances as well, such as, for example, Persistent Organic Pollutants, radioactive substances or pollens.

1 Introduction

The rapid rise in automobile ownership and gasoline consumption in the early 1970s, leading to an estimated 375 000 tons of lead per year worldwide (Nriagu, 1992), coincided with the period when scientific evidence was uncovering some of the effects of lead as a neurotoxin (reviewed by Lovei, 1997). Even at low exposure levels, the effects on children's development and health include the impairment of normal neurological development leading to learning and reasoning difficulties, the retardation of physical development, hearing loss, hyperactivity, and reduced attention span. Effects in adults include elevated blood pressure and hypertension, resulting in an increased risk of cardiovascular diseases, and renal deficiencies. Health concerns led to the prohibition in the 1970s, in developed countries worldwide, of lead-based paint, and of the usage of lead solder in water pipes and in food cans. Gasoline lead (tetraethyl and tetramethyl lead), added for its anti-knock properties (at 0.6 g Pb/l in Europe), and representing by far the largest source of lead in the atmosphere, was to become the next target of European regulations. After the maximum allowed gasoline lead content was reduced in the 1970s, the appearance of a car engine catalyst that reduced automobile emissions of NO_x, CO and C_xH_y but was inhibited by lead resulted in the introduction of unleaded gasoline. Finally, the 1998 Aarhus Treaty stipulates the exclusive usage of unleaded gasoline in Europe by the year 2005.

In the "lead study" conducted at the Institute for Coastal Research at GKSS Research Center, Germany, the case of gasoline lead emissions and regulations in Europe was analyzed. Specific questions asked were: How did lead emissions, atmospheric concentrations and depositions develop since the 1950s? Was the decline in air concentrations of lead matched by corresponding declines in plants, animals and humans? Did gasoline-lead regulations result in considerable economic burdens in Germany (the main promoter of such policies)? To address these questions, we use a combination of computer-model simulations and measurement data to reconstruct atmospheric lead transport and deposition since 1958. Based on lead emissions estimates, the simulation models reconstruct the evolution over time of the spatial patterns (at about 50 km resolution) of environmental lead levels in the 38-year period 1958-1995. This evolution exhibits a sharp rise leading up to the mid-1970s followed by an even more dramatic fall to below 1958 levels. Some ecological and health effects of this evolution are analyzed,

including a fall by two-thirds at some locations in the lead levels in plant leaves as well as in human blood, in the period from the mid-1970s to the mid-1990s. Economic impacts of German regulations on this country's industrial markets are also explored. We conclude that regulations were mostly successful in bringing down high environmental lead levels, while they did not result in undue burden to the German economy.

A methodological result of this study is that long-term reconstructions of regional environmental change can be achieved with dynamical regional environmental models, including climate and tracer transport. Past political and economic evolutions may be assessed by a retrospective analysis. By implication, scenarios of environmental impact of forecasted socio-economic developments are also possible to be constructed.

In this paper, we first review the history of gasoline lead content regulations in Europe (Section 2) and the history of lead emissions, of which the major source is road traffic (Section 3). In Section 4 the results of our main effort are presented, namely the detailed modelling of the atmospheric pathways and depositions of lead throughout Europe form 1958 until 1995 with a spatial resolution of 50 km and a temporal resolution of 6 hours. The effect of regulations on plants, animals and humans is considered in Section 5, and their impact on German industrial markets is dealt with in Section 6. Conclusions and perspectives for future studies are presented in Section 7.

2 The history of European gasoline lead content regulations

The history of European lead content regulations and the role played by the German government in promoting such regulations was reviewed by Hagner (2000) and is summarized here.

Environmental matters in the early 1970s weighed especially heavily in German politics (Peters, 1980), and Germany would be the first European country to impose restrictions on the allowed lead content in gasoline. Starting in 1972, German production and importation of gasoline with over 0.4 g Pb/l was prohibited (down from the usual 0.6 g Pb/l), and starting in 1976 the more strict limit of 0.15 g Pb/l was imposed. In contrast, the European Union fixed its limit modestly at 0.4 g Pb/l starting only in 1981, and stipulated that no individual country was allowed to set a limit lower than 0.15 g Pb/l (Council Directive 78/611/EEC of

1978).

In the early 1980s, eliminating lead altogether from gasoline became an attractive possibility because a new combustion catalyst was introduced which reduced the emissions of NO_x, CO and C_xH_y but which could operate only at very low lead concentrations. These gases were mass pollutants believed to pose a threat to forests through acidification and photo-oxidation. The German government wanted to adopt the new catalyst and introduce car-emission regulations as strict as those already in place in the US and Japan (Deutscher Bundestag, 1984), invoking grave concern for its national forests.

Unleaded gasoline (0.013 g Pb/l) was introduced in Germany in October 1984. To prohibit the sale of leaded gasoline in Germany was not an option because the European Union dissallowed 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 (see Section 6).

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 new lead-averse catalyst). In 1985, the EU mandated that by October 1989 Super unleaded gasoline be available for sale in all member states, alongside the leaded variety (Council Directive 85/210/EEC). Moreover, member states were asked to voluntarily adopt a 0.15 g Pb/l limit. Unleaded gasoline was defined as containing no more than 0.013 g Pb/l. In 1987, Council Directive 87/416/EEC emphasized the importance of the availability of unleaded gasoline for sale in every country. All member states were then allowed to prohibit national production and sales of leaded 92-octane gasoline. Observable damage to public health and the environment was invoked (Rat der Europäischen Gemeinschaften, 1987).

While adherence to unleaded gasoline was quite prompt in Austria, Denmark, Germany, The Netherlands and Sweden, other countries initiated their phase-out only much later. France offered strong resistance, partly to protect its small-car export industry. The responses by the different EU countries were reviewed by Hagner (2000) and Lundberg et al. (submitted) (see

Section 6). By 1995, unleaded gasoline had conquered over 80% of the market in Austria, Finland, Denmark, Germany and The Netherlands, but only under 30% in Italy, Greece and Portugal. Lunderg et al. (submitted) identify higher leaded-gasoline prices and the widespread adoption of cars using the lead-averse catalysts as the two most effective factors in reducing the market share of leaded gasoline.

The Aarhus Treaty, signed in 1998 by all Western and nearly all Eastern European countries, stipulates the exclusive usage of unleaded gasoline by the year 2005 (COWI and DTI, 1998).

Also fixed-source emissions (industrial and others) have significantly decreased since the mid 1970s.

3 Reconstructed lead emissions

Pacyna and Pacyna (2000) provided expert estimates of European atmospheric lead emissions for the years 1955, 1965, 1975, 1985, 1990 and 1995, and projection estimates for the year 2010. Pie charts of emissions by source category and by country are given in Figures 1 and 2, respectively, and the data are provided in Table 1.

Total European atmospheric lead emissions have changed dramatically in the course of these four decades, but road transport has consistently remained by far the largest emission source throughout the period (Figure 1). Road lead emissions totaled an estimated 31 thousand tones (tt) in 1955 and nearly quadrupled to 119 tt in 1975 with the motorization of the masses. While road transport and gasoline consumption continued to rise, subsequent gasoline-lead content regulations nearly halved road lead emissions to 62 tt in 1985; and, as unleaded gasoline conquered increasingly higher market shares, road lead emissions further dropped to 42 tt in 1990 and to 19.5 tt in 1995. The forecast for year 2010 is a comparatively small 7.6 tt.

Despite the abatement of lead emitted by road transport, this source category remained the largest by far. This was possible because lead emissions by other source categories suffered comparable or even more dramatic drops, in response to various economic, technological and process changes. Thus, road transport accounted for about one-half of total emissions in 1955, about three-quarters from the mid-1970's through the mid-1980's, and over two-thirds in 1995. Despite the 1998 Aarhus Treaty, stipulating the exclusive usage of unleaded gasoline (which

has the smaller lead content of 0.013 g/l) in Europe by the year 2005, the projected emission estimates still predict road transport to remain as the major lead source, accounting for about 60% of total lead emissions in year 2010.

The European portion of the former USSR has been the largest lead emitter in Europe throughout the estimation period, and is predicted to maintain that position in year 2010 (Figure 2). Together, Russia and Ukraine accounted for over one-half of European lead emissions in 1995, a fraction they are predicted to maintain in 2010, largely due to their continued usage of leaded gasoline. In 1995, about 80% of Russia's and nearly 70% of Ukraine's total atmospheric lead emissions were generated by road transport (amounting to an estimated 8822 and 2332 tones, respectively). In these countries, the major part of gasoline produced today is low-octane. Because the demand for high-octane gasoline is increasing, these countries face the added challenge of controlling the quality of their gasoline because lead is often added to gasoline after it has left the refinery, to increase the octane rating (Lundberg et al., submitted).

Other major lead sources in Russia and Ukraine in 1995 were associated with iron and steel production (generating an estimated total of 1345 tones in the two countries) and, in Russia, non-ferrous metal manufacturing (1080 tones). Lead emissions by iron and steel production and by non-ferrous metal manufacturing had also been high in several Western-European countries in the past, but by 1985 had already been drastically reduced. Western Germany (FRG), for example, had emitted an estimated 3900 tones of lead from non-ferrous metal manufacturing and 1500 tones from iron and steel production in 1965, but by 1985 had reduced these numbers to 170 and 330 tones, respectively. The largest emitters in Western Europe throughout these four decades have been Germany, France, the U.K. and Italy, with Germany showing the most dramatic abatement in the 1980s and 1990s.

In order to be used by our atmospheric transport simulation model (Section 4), lead emission estimates had to be converted into gridded form (Figure 3). To assign emissions to grid cells, the geographical coordinates of stationary sources were used. For road traffic, emission estimates were available at the country scale only, and were disaggregated using data on population density in each grid cell. Gridded emissions for intermediate years were obtained by linear interpolation between the emission values in the two nearest years of estimate. As a result, any intermediate emission peaks in local estimates are not captured. The highest lead

emissions peak in the four decades occurred in the vicinity of 1970 in many countries, while our emission estimates for 1970, equal to the average of 1965 and 1975 estimates, are likely to be underestimated in those countries. Emission rates were held constant within each year. Thus, any seasonal or diurnal emission variability is not reproduced.

4 Reconstructed regional weather and atmospheric transport and deposition of emitted lead

In this section we describe the methods and results of the computer simulations performed to reconstruct the regional weather conditions and the pathways of atmospheric transport of emitted lead and its deposition from the atmosphere to the earth surface.

The climatic conditions over Europe during the 40-year period 1958-1998 were reconstructed by the global "reanalyses" of the US National Centers for Environmental Prediction (NCEP) (Kalnay et al., 1996), at about 2° spherical resolution. The NCEP reanalyses are in accord with the available observations, and are sometimes referred to as "observed states".

While large-scale reanalyses features can be accepted with confidence, their finer-scale features may not be equally well represented. 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 of the technique known as "regionalisation" is to retain the large-scale features while adding the regional detail related to physiography.

We performed a regionalisation of the NCEP reanalyses features, yielding a 41-year (1958-1998) dataset with 0.5° spherical resolution and 1 hour temporal resolution (Feser et al., 2001). This regionalisation was performed with the regional atmospheric model REMO (Jacob and Podzun, 1997). The regional model was exposed to the global coarse grid analyses not only within a domain-boundary sponge region (as is conventionally done) but also in the interior of the domain, using the technique of "spectral nudging" (von Storch et al., 2000).

Comparisons with local observations showed that the quality of the data set was favorable and uniform in time (see Feser et al., 2001). The data-set is considered to be of good quality, suitable for being used in various applications, and are made available for public use through the German Climate Computing Center (DKRZ).

Airborne lead is attached to particulate material, which can travel in the atmosphere even to remote areas (Hong et al., 1994; Rosman et al, 1995). To simulate the transport of lead-carrying particles in the atmosphere we used the TUBES model (Costa-Cabral, 1999, 2001), using as input (or "forcing") the REMO weather reconstruction, and with computational schemes for dry and wet deposition similar to those of the EMEP/GKSS model (Petersen et al., 1989; Krüger, 1996). A particularity of this model compared to other horizontal Lagrangian models is that it explicitly takes into account the horizontal con- and divergence of the wind field by using flow tubes instead of linear trajectories, so that the plume of a transported substance may become wider and narrower. This scheme allows for mass conservation at any scale.

Dry deposition is the removal of the lead-carrying particles from the atmosphere to the earth surface, in the absence of precipitation. The rate of dry deposition is assumed to be proportional to the lead mass in the air column, m. The "dry deposition velocity," v, is the proportionality constant. Wet deposition is the removal by precipitation. The rate of wet deposition depends linearly on the precipitation rate, p, and on its "scavenging efficiency," w. We assume w to be constant and equal for both rainfall and snowfall. Following the results of Petersen et al. (1989), we used $v = 0.2cm/s$ and $w = 500000$. As an emitted mass of lead travels downwind in a flow tube, it is progressively depleted over time by dry and wet deposition. A mathematical description of deposition in TUBES is given in Costa-Cabral (2001).

As input to the TUBES model (model "forcing"), we used the estimated lead emission rates in gridded format (Section 3); the horizontal wind field at the 925 hPa pressure level (roughly, 800 m altitude), sampled at 6-hour intervals (the 925 hPa level horizontal wind fields are considered representative of the major horizontal advective transport directions in the mixing layer (Petersen et al., 1989)); the precipitation rate (rain and snow) (Figure 4), which was provided hourly by REMO and was integrated over 6-hour intervals to drive wet deposition; and the depth of the mixing layer, computed for each grid cell at 6-hour time intervals from several variables (the variables required for this computation are atmospheric pressure, specific humidity, potential temperature and height of the REMO vertical layers; a combination of the "dry parcel" and "Blackadar" methods was used).

Figures 5 and 6 display the simulated mean annual lead air concentration and deposition rates for selected years over the domain. Comparison. (In this comparison note, however, that

the first emissions map shown corresponds to 1955 (the first year of emissions estimate), while the first concentrations and depositions map corresponds to 1958 (the first year of simulation).) of each of these two figures with the emission rates in Figure 3 shows that spatial patterns of concentrations and depositions over land areas roughly follow the pattern of emission rates, decreasing over marine areas with distance to the coast. Like the emissions, also the concentrations and depositions rose in the period before 1975, after which they began to fall. (1975 is a year of estimate for emission rates. The closest other years of reference are 1965 and 1975. Therefore, the true peak in emissions and concentrations may have occurred not in 1975 but in some other year between 1965 and 1985.)

How do lead emissions from one country affect the air concentrations and deposition rates in nearby countries and marine regions (see Figure 7)? Table 2 displays the "average contributing matrix" for countries and marine regions, that is, the fraction of the deposition in a given country or marine region (listed in columns) originating in a given country (listed in rows), averaged over the 38-year simulation period. (Emissions from islands in the North Sea and Baltic Sea were, for the purposes of these calculations, considered to be marine sources, that is, their emissions were not assigned to the country to which the islands belong politically.)

Most countries listed in Table 2 are large in size. As a result, the overwhelmingly largest fractions correspond to contributions from a country to itself (diagonal-row values). Some of the smaller countries in the table however have substantial fractional contributions from neighboring countries. Of the lead deposited over Switzerland in the 38-year simulation period, an estimated 21% originated in France, 12% in Italy, and 10% in Germany. Similarly, about one-fifth of the lead deposited in the Netherlands originated in Germany, and another fifth in Belgium. About a quarter of all lead deposited in Ireland was emitted from the U.K.

Year-to-year variability in contribution fractions reflects meteorological variability, that is, changing patterns of wind direction and precipitation rates. It also reflects emission changes, but to a lesser degree, given that in these simulations emissions changes are gradual and smooth from one year to the next as a result of being obtained by interpolation (as noted above). Figures 8a and b illustrate contribution fraction variability for the receptor countries Switzerland and Ireland. For Switzerland (Figure 8a), note how the contributions by France and Italy to some extent approximate mirror-images of each-other. Years in which southerly

winds are more frequent over this region — such as 1973, for example — favor higher Italian and lower French contributions. The variability in U.K. contribution to Ireland (which ranges from 0.16 to 0.33) and Ireland to itself (Figure 8b) are higher than in the case of Switzerland because only two countries are involved, and because of the large difference in emission rates between these two countries.

Next we will look at simulation results for selected marine and land areas.

Baltic Sea and North Sea

Atmospheric substance inputs to the Baltic Sea are of particular concern due to the propensity for accumulation in this sea's sediments, long mean water residence time — estimated to be between 20 and 30 years —, and from its shallow mean depth of just 52 m. The long residence time is due to limited exchanges with the North Sea. The atmosphere represents the largest source of lead and other heavy metals to the Baltic Sea (e.g., Schneider et al., 1993, 2000).

Figure 9 shows the simulated total annual lead deposition to the Baltic Sea area throughout the 38-year period. Also indicated in Figure 9 are estimates by different authors based on measurements, as reported in Schneider et al. (2000). According to Schneider et al. (2000), the disagreement between some of these measurement-based estimates may be due to inadequate sampling and analytical techniques, and to using different methods for extrapolating from coastal data to the entire Baltic Sea. An overall agreement is seen between the simulation results and the measurement-based estimates, which is particularly good for the Rodhe et al. (1980) and the HELCOM (1989; 1991) estimates.

Figure 10 shows the fraction of contribution by individual countries to deposition in the Baltic Sea. A country's contribution depends on the strength of its lead emissions, its distance to the Baltic Sea, its directional position relative to the Baltic Sea given the predominant wind directions, and precipitation patterns. Southwesterly winds predominate in the region, hence sources located in the southwestern vicinity of the Baltic Sea contribute disproportionately.

Figure 11 shows the simulated total annual lead deposition to the North Sea area throughout the 38-year period. Simulated depositions are much lower for the North Sea than for the Baltic Sea, by a factor of roughly two to three throughout the period, despite the former's larger surface area. This is explained by the northwesterly winds that prevail over the North Sea

region and reduce the contributions from Continental Europe. Deposition over the North Sea shows an extended peak covering the 1965-1975 period, appearing earlier than the Baltic Sea peak of 1974 (Figure 9). The earlier peak over the North Sea is explained by that emissions by the U.K., its main contributor, peaked in 1965 and decreased only slightly from 1965 to 1975, while the three main contributors to the Baltic Sea — the Russian Federation, Germany and Poland — increased their emissions from 1965 to 1975 (Table 1). The marked year-to-year variability in Figure 11 is due to meteorological factors.

The distribution of simulated air concentrations and depositions in the North Sea and Baltic Sea for selected years can be seen in Figures 6 and 7, respectively. It can be better seen in the larger-scale Figure 12 for the last year of simulation, 1995. The location of EMEP measurement stations on coastal areas of these two seas is also indicated in Figure 12. Note the rapid gradient in concentrations along these coastal marine areas, where the simulated value may differ greatly between adjacent grid cells.

The EMEP stations (Figure 12) are located on the coast, where the strongest concentration gradients lie, and where any given point location such as an EMEP station is not representative of a grid cell. Nevertheless, fairly good comparisons are obtained between model grid-cell values and EMEP-station measurements (EMEP, 1996, 1997, 1998), reported in Table 3 and displayed in Figures 13 and 14.

We see that the northerly station measurements GB91 and NO99 (for which the lowest values among all coastal stations are reported) are consistently under-estimated by the simulations. Under-estimation is obtained also for the Belgian station BE90 (for which the highest measured values of all coastal stations are reported: 104.2 and 90.0 ng m^{-3}, respectively). The values observed at these three under-estimated stations are however generally reproduced by the simulations at only short distance inland, indicating that the under-estimation may result from lack of sufficient spatial resolution by the model simulation over these high-gradient coastal areas.

Storelungmose, Denmark

The deposition flux of lead from the atmosphere to the ground at Storelung Mose (Staaby, Denmark) was estimated by Goodsite et al. (2000; 2001) using measurements of lead content

and dating techniques for peat core slices. Figure 15 shows the curve of measurement-based estimates which, extending from 1902 to 1997 nearly covers the entire 20th century; and the shorter curve of 1958-1995 simulated fluxes. The agreement between these curves is remarkable.

Recall from Section 2 that emission rates were estimated for 1955, 1965, 1975, 1985, 1990 and 1995, and the emission rates used for intermediate years were obtained by linear interpolation between the two closest years of estimate. The considerable year-to-year variability exhibited by the simulation curve is therefore not explained by emissions variability alone, but is largely due to meteorological variability.

Recall also that, in many countries, the highest emission peak is believed to have occurred in the vicinity of 1970 (Section 2). Because the emission rates for years between the estimate-years 1965 and 1975 were obtained by linear interpolation, then it is likely that simulated deposition rates (and concentrations) are under-estimated for some of the years in the 1965-1975 period. This means that a peak occurring between 1965 and 1975 may be "missing" from the estimation curve.

Central and East-Central Europe

Figure 16 displays the observed versus simulated lead air concentrations at the Central and East-Central European EMEP stations, for each year from 1988 to 1995. Measurements for Southern Europe are extremely scarce and do not allow proper comparison with estimates.

Simulated values are generally within a factor of 2 from the observed values (Figure 16). When averaged over the 8 years, simulated values for all stations are within a factor of 2 from the observed 8-year means. There appears to be a tendency for over-estimation in the stations of Germany, Poland, Latvia and Estonia, but a tendency towards under-estimation in the stations in the Czech Republic.

5 Some effects on human, plant and animal populations

Upon deposition lead accumulates in the soil and plants, and enter the food chain through which it may reach human populations. What have the effects been of the rise and fall of atmospheric lead concentrations and deposition rates on lead levels on plants, animals and

people?

Terrestrial plants, whose capability to uptake and accumulate heavy metals varies widely between species (König and Krämer, 1985), take lead in mostly from the atmosphere rather than the soil (Chamberlain, 1983). This explains why lead levels in plant leaves, needles and sprouts were found to be well correlated with atmospheric concentrations for each given plant species. For example, lead concentration in poplar leaves in urban areas in Spain were shown to correlate positively with local traffic density (Toro et al., 1995).

Atmospheric lead uptake is most important for foliage plants (König, 1986). Uptake increases with leaf area and leaf surface roughness, and depends on plant age. Schultz (1987) reported that the upper canopy layer of spruce forests retained over half of the deposited lead. Despite the influence that species, location and seasonal factors have on lead capture and intake by plants, a clear declining trend nevertheless emerged in the lead concentrations of leaves, needles and sprouts documented by various studies. Figure 17 shows the evolution from 1985 to 1996 of lead concentrations in sprouts of annual spruce (*Picea abies*) and leaves of poplars (*Populus nigra*) in the German state Saarland (Umweltprobenbank, 1999). This decay is fully consistent with the modelled drop of atmospheric lead concentrations (Figure 5).

In contrast with terrestrial plants, aquatic organisms are mainly influenced by the long-term accumulation of lead in fluvial and marine sediments. For example, Hoebel (1984) found the lead concentration in foraminifers to be correlated with that in their local surrounding sediments. In terrestrial soils lead accumulates mostly in the humus layer, compounded in metalo-organic complexes. Particles found in fluvial and marine systems with high lead concentrations are mostly not the result of atmospheric deposition but of fluvial discharge sources instead (Hagner, 2002). Additionally, sediments act as a sink for lead, which is released from them only very slowly. Therefore, the decreasing atmospheric lead deposition in the 1980s and 1990s has hardly influenced lead loads in aquatic systems. As an example, Figure 18 shows the evolution from 1982 to 1997 of lead concentration in blue mussels (*Mytilus edulis*) along the German North Sea coast. No clear decreasing trends are demonstrated. A similar conclusion is obtained from the analysis of muscle tissue of fish in the river Elbe (see Hagner, 2002).

The German Human Biomonitoring Commission offered in 1995 categories for the risk associated with different lead concentrations in human blood. Levels above 250 μg Pb/l were

considered risky for adults, requiring medical analysis. For children and embryos, levels of 150 μg Pb/l were declared critical. In the United States some researchers are convinced that the intellectual development of children is disturbed already at a blood lead level of 100 μ g Pb/l (CDC, 1991). Figure 19 displays lead concentrations in human blood in Germany based on various different samples, and a continuous series of analyses among students from the University of Münster. Measurements represented by triangles and circles were taken at different German locations and used different methods. A significant drop in blood lead concentrations since 1979 is apparent from the figure, even if a secondary maximum emerged in about 1988. During this time, the lead concentrations were under the critical level of 150 μg Pb/l, but if the US researchers are right, then there may have been health impacts on children in the early 1980s.

One study where all blood samples were collected at nearby locations and uniform methods were used is that of Krause et al. (1996), which therefore allows comparison between genders and different age groups. For children aged 6 and 7, they report an average of 38.8 ug Pb/l in blood, while for adults the concentration increases with age from an average 44.2 ug Pb/l at 25.29 years of age to 55.8 ug Pb/l at 50-59 years of age. Differences between genders were negligible.

Unfortunately, no blood lead data prior to 1979 is available to us, so that nothing definite can be said about its concentrations during the air concentration peak of the early 1970s. Therefore, we have constructed a simple linear relationship between modelled air concentrations in Münster (Germany) and the Münster blood level series (Figure 19; dashed line). According to this admittedly crude estimate, maximum mean levels may have been about 150 μg Pb/l. If this estimate is realistic, then some children born around this time may have been at some risk. One has to keep in mind that the numbers given are sample means, so that concentrations in some individuals would have been higher, while in others lower.

6 Economic effects in German industrial markets

Effects of gasoline lead content regulations on the German mineral oil and automobile industries were studied by Hagner (2000) and are summarized here.

Mineral oil industry: refineries and gas stations

Prior to the first reduction of gasoline lead content in 1972, the mineral oil industry had argued against the measure, voicing concerns about additional costs. However, instead of rising costs actually dropped, thanks to savings with reduced lead additives. It was only after the second regulation in 1976 that fuel production costs indeed rose, because new additives with high octane numbers were needed for maintaining gasoline performance.

In the course of time, concerns about a sufficient supply were voiced, but none of the anticipated bottlenecks in gasoline supply took place. The German refinery sector had built up large overcapacities so that in 1976 only 60% of total capacity was in use. In 1985, the mineral oil industry had already built new conversion production plants which could produce gasoline with high octane numbers without lead additives (Hagner 2000).

From 1950 to 1992, gasoline sales in Germany increased substantially (Figure 20). This development, however, was interrupted by the oil crises in 1973 and 1979 which triggered two worldwide recessions and drops in gasoline sales. Due to the lead reduction regulations in 1972 and 1976, the total lead emissions decreased significantly (Figure 20). Despite the implementation of unleaded gasoline in the autumn of 1985, lead emissions decreased only slowly in the following years because of unleaded gas was only slowly accepted by the consumer.

Several factors negatively influenced the introduction of unleaded gasoline. Initially, it was more expensive than leaded gasoline (a 0.1 DM/l difference). In early 1986 the price became nearly the same. From early 1988 on, however, unleaded gas became about 0.1 DM/l cheaper than leaded gas. This change was achieved by tax subsidies. Furthermore, adaptation measures in the automobile motor had to be implemented because quality norms were different for leaded and unleaded super gasoline. The minimum octane number was 85 and 88 for leaded and unleaded gasoline, respectively. To accomodate this difference, valves had to be redesigned (Umweltbundesamt, 1980). Additionally, the media argued against the use of unleaded fuel (IPOS, 1986). It was claimed that unleaded gasoline contained a high percentage of carcinogenic benzene and that gasoline consumption would rise. The negative attitude was shared by many of the automobile garages and petrol stations, who fearing significant investment costs (Westheide, 1987).

Reorganization at gasoline stations was required in order to add an extra pump for the new unleaded variety. Many stations, in particular the smaller ones operated by independent traders, had to build new gasoline pumps and containers, while the larger stations operated or franchised by companies often had only to reassign their pumps. Furthermore, until late 1987 the supply of unleaded gasoline was difficult for the independent trade (Statistisches Bundesamt, 1987). The traders could receive unleaded fuel only from domestic refineries, which sold it for the same price as the petrol stations owned by large companies. Although the German government implemented investment subsidies for these middle-class firms, the independent petrol stations lost some of their market share. In 1985, they held 11% of the market of leaded gasoline, but only 4.5% of the market of unleaded gasoline (Westheide, 1987).

Automobile industry

While the gasoline-lead regulations of the 1970s had little effect on the German automobile industry, the introduction of catalytic converters and unleaded gasoline in the 1980s implied additional costs - between 1500 and 2500 DM (750 to 1250 Euro) per car. Over time, the costs for the converters declined, thanks to mass production and strong price competition. During the first years after the introduction of catalysts, Daimler-Benz and VW were the major winners among German car manufacturers (Monopolkommission 1987). Daimler-Benz produced engines that were easily compatible with catalysts and customer demand was relatively insensitive to price changes. VW benefited because of its large array of catalyst-equiped cars. Market share losses were suffered especially by Renault, Opel-Deutschland and Ford-Deutschland. The market shares for cars with and without catalytic converters differed strongly. For Renault it was 3.5%:2% (without converter : with converter), for Ford-Deutschland: 12.1%:8.2% and for Opel-Deutschland 15.4%:9.8%. Mercedes-Benz and Volkswagen, on the other had had edges of 10.7%:17.7% and 21.6%:28.7% (Westheide (1987). Overall, favourable terms of competition were experienced by producers of cars with high technical standards, who had already gathered experience with catalyst systems on the U.S.-market (Hagner 2000).

7 Conclusions

The atmosphere and the environment in general, will remain for the foreseeable future a dump for various anthropogenic substances. Some of these substances will have negative properties so that society will sooner or later begin regulating their emissions. To this end, science has to provide society with the tools to assess the situation in the past and to evaluate the possible impacts regulations may have. We have developed such a tool, made up of a detailed emission chronology, a regionalized history of weather events and an atmospheric transport model. In the future this tool needs to be completed by a model describing the transport, transformations and depositions in catchments and rivers.

This tool has been applied to airborne lead originating mostly from road traffic. We have chosen lead for two reasons. First, lead emissions underwent significant changes, from an almost unabated increase to a series of sometimes drastic reductions. Thus, there is a well defined signal to be detected and described. Second, lead behaves during its aerial transport to first order approximation inert, so that the simulation of transport and deposition is relatively simple, compared to more reactive substances such as mercury or persistent organic pollutants (POPs).

We have demonstrated that for the case of lead our tool is functioning well. It remains to be seen if the success can be repeated with other airborne substances, such as radioactive substances, POPs, or pollen.

Our modelled data show that lead concentrations increased heavily until the 1970s. Later, the atmospheric concentrations fell strongly, quite consistently with the sparse observational evidence, mainly because of the reduced lead content in gasoline. As a consequence the lead levels in organisms declined for those that take in lead from the atmosphere like terrestrial plants and humans. But in aquatic organisms the lead concentrations remained at a constant level. The economic impacts of the regulations were small in Germany. Except for some short-term effects it hardly burdened the German economy. Other European economies have not been studied here.

Gasoline lead reduction regulations in Europe may be considered a successful example of environmental policy. However, the success of those policies in protecting biological systems from

lead exposure was limited to atmospheric pathways, underscoring the fact that a short residence time is a necessary condition for substance abatement through emission regulations in a given environmental compartment once considerable substance amounts have already been released. For those anthropogenic substances that persist for years, that are subject to bio-acumulation, and whose main route of human exposure is the food chain, late emission regulations may be ineffective in protecting human health. In such cases, the principle of prevention, by which any significant releases should be precluded from the start, may be indicated.

One should, however, not forget that the large amounts of lead emitted in the past 50 years have not simply vanished but reside now for good ubiquitously in the environment. The use of lead in gasoline was indeed a large-scale geophysical pollution exercise, and it remains to be seen if some long-term effects may appear at a later time. Also this insight may serve as a valuable lesson from the study of the history of lead emissions.

Acknowledgements. The help of Beate Gardeike in preparing some of the figures is gratefully acknowledged. The project was funded by the Helmholtz Foundation (HGF), Germany. The lead project web page: <http://w3g.gkss.de/staff/blei>.

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Tables and Figures

Table 1: Anthropogenic emissions of Pb to the atmosphere in Europe during the period 1955–2010.
Source: Pacyna and Pacyna (2000).

Country	1955	1965	1975	1985	1990	1995	2010
Albania	61.2	124.6	219.8	150.0	113.0	33.4	11.1
Austria	1892.5	2749.7	2618.6	790.0	215.4	39.3	33.7
Belarus	– (1)	– (1)	– (1)	– (1)	– (1)	47.0	20.3
Belgium	3603.1	4789.1	4236.1	1490.0	577.0	435.0	105.8
Bos.-Herz.	– (2)	– (2)	– (2)	– (2)	– (2)	38.6	22.9
Bulgaria	705.3	1435.1	2531.4	1590.0	1397.0	297.5	102.7
Croatia	– (2)	– (2)	– (2)	– (2)	– (2)	286.0	144.6
Czech Rep.	941.9 (3)	1525.8 (3)	2914.3 (3)	1170.0 (3)	577.6	376.6	116.2
Denmark	1101.0	1599.8	1523.5	300.0	163.2	16.2	11.9
Estonia	– (1)	– (1)	– (1)	– (1)	– (1)	58.0	34.5
Finland	923.4	2056.1	2964.7	930.0	326.5	67.0	49.4
France	7377.6	12968.9	18862.5	8610.0	2987.0	1510.5	504.2
Germany	–	–	–	–	4074.0	624.0	328.7
FRG	9349.1	13583.8	12936.3	4617.0			
DDR	875.4	1386.3	3509.2	1870.0			
Greece	322.0	1066.6	1874.9	790.0	436.0	324.0	113.4
Hungary	268.3	545.9	962.9	666.0	604.0	153.7	63.4
Iceland	–	–	–	–	15.0	4.0	3.3
Ireland	439.3	523.2	518.1	390.0	213.0	85.2	50.8
Italy	2383.9	7897.4	13882.4	5490.0	2861.0	2174.0	561.5
Latvia	– (1)	– (1)	– (1)	– (1)	– (1)	10.3	4.4
Lithuania	– (1)	– (1)	– (1)	– (1)	– (1)	19.4	8.4
Luxembourg	315.4	457.4	416.9	160.0	100.0	29.8	13.3
Macedonia	– (2)	– (2)	– (2)	– (2)	– (2)	63.3	37.6
Moldavia	– (1)	– (1)	– (1)	– (1)	– (1)	23.0	13.7
Netherlands	1109.6	1956.2	2263.7	1341.6	266.0	152.0	71.9
Norway	598.3	1332.2	1920.8	412.0	162.3	28.4	27.7
Poland	2442.1	4225.8	6981.9	3000.0	1441.7	959.7	352.7
Portugal	88.0	291.6	512.6	390.0	209.0	209.0	46.3
Romania	519.0	1056.0	1862.6	1420.0	1423.0	937.5	419.0
Russia	14558.0 (1)	31150.5 (1)	54131.5 (1)	30500.0 (1)	30457 (1)	11000.0	4742.4
Slovakia	– (3)	– (3)	– (3)	– (3)	331.2	97.0	57.1
Slovenia	– (2)	– (2)	– (2)	– (2)	– (2)	195.0	51.6
Spain	1659.8	2705.9	6325.0	3620.0	2435.0	1826.0	1020.9
Sweden	851.0	1895.0	1600.0	950.0	365.0	37.8	36.6
Switzerland	423.0	941.9	1358.1	480.0	248.0	226.0	99.2
Ukraine	– (1)	– (1)	– (1)	– (1)	– (1)	3400.0	1910.2
U.K.	8841.7	10528.9	9140.3	8114.5	3795.1	1541.0	784.5
Yugoslavia	881.8 (2)	1794.2 (2)	3164.9 (2)	2340.0 (2)	2337.0 (2)	1065.0	632.2
Total	62531.7	110587.9	159233.0	81581.1	58130.0	28390.2	12608.1

1) former USSR
2) former Yugoslavia
3) former Czechoslovakia

Table 2: Average "contributing matrix" for selected countries and marine regions. The matrix is composed of the average fraction of the deposition in a given receptor country or region (listed in columns) originating in a given emitting country (listed in rows). Only those fractions larger than 0.01 (1%) are displayed. Fractions exceeding 0.1 (or 10%) are highlighted in red.

	Spain	France	Switz.	Italy	Austria	Germany	Belgium	Nether.	U.K.	Ireland	Norway	Denmark	Sweden	Finland	Poland	Russian Fed.	North Sea	Baltic Sea	Med. Sea	West-Atlantic
Spain	0.94	0.01																	0.11	0.23
France	0.04	0.89	0.21	0.02		0.10	0.25	0.06	0.05	0.01							0.24		0.08	0.23
Switz.		0.01	0.54	0.01	0.01	0.01													0.37	
Italy		0.01	0.12	0.87	0.06															
Austria			0.02	0.01	0.62	0.02														
Germany		0.02	0.10		0.17	0.70	0.09	0.20				0.12	0.01		0.10		0.09	0.20		
Belgium		0.02				0.07	0.57	0.21	0.01								0.07			
Nether.						0.03	0.04	0.48				0.01					0.06			
U.K.		0.01					0.02	0.03	0.90	0.24							0.34			0.27
Ireland									0.01	0.68							0.01			0.05
Norway											0.76		0.05	0.02			0.01	0.01		
Denmark											0.01	0.58	0.02				0.04	0.05		
Sweden											0.10	0.01	0.76	0.02				0.12		
Finland											0.05		0.04	0.67	0.04	0.01		0.16		
Poland					0.02	0.05									0.76			0.23		
Russian F.											0.01			0.16		0.91		0.09		

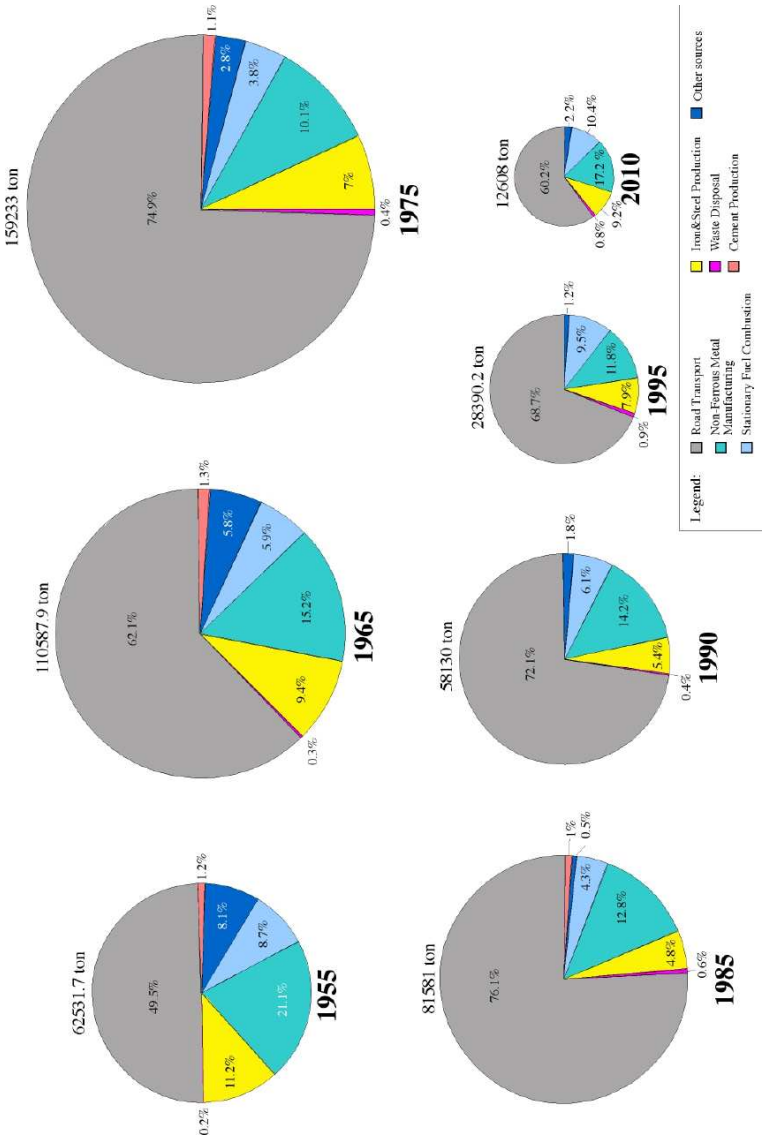

Figure 1: Pie charts of atmospheric lead emission percentage by source category in each year of estimate (data from Pacyna and Pacyna, 2000).

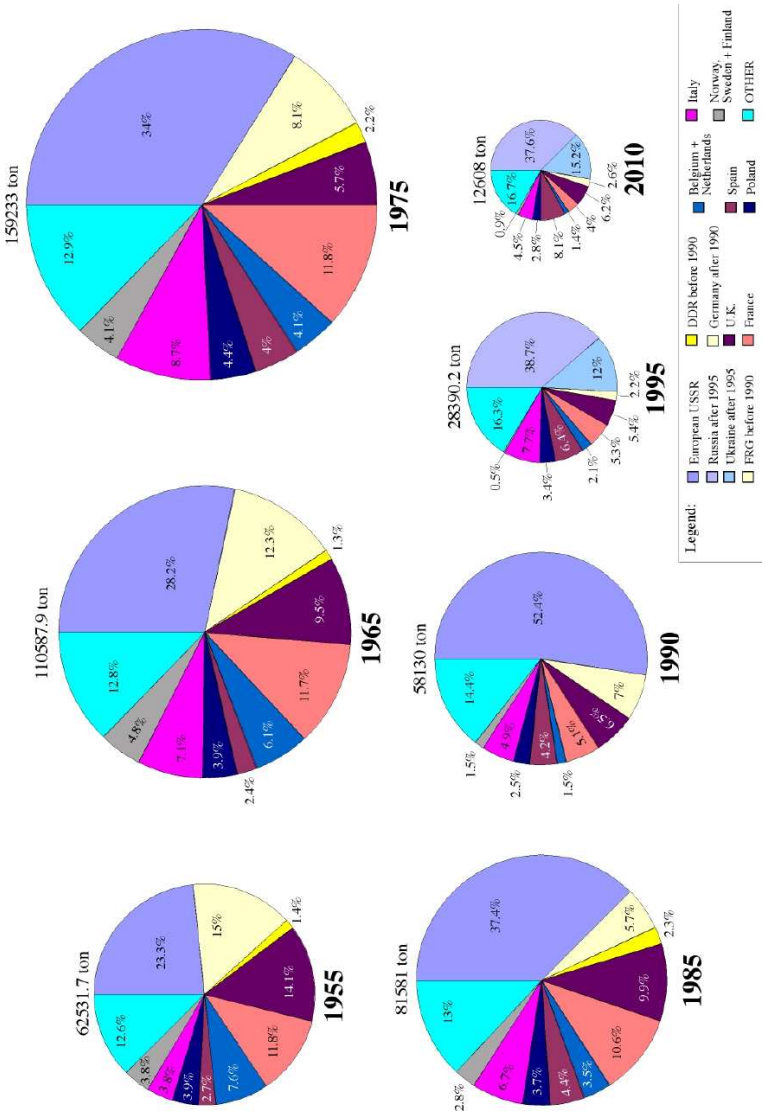

Figure 2: Pie charts of atmospheric lead emission percentage by country in each year of estimate (data from Pacyna and Pacyna, 2000).

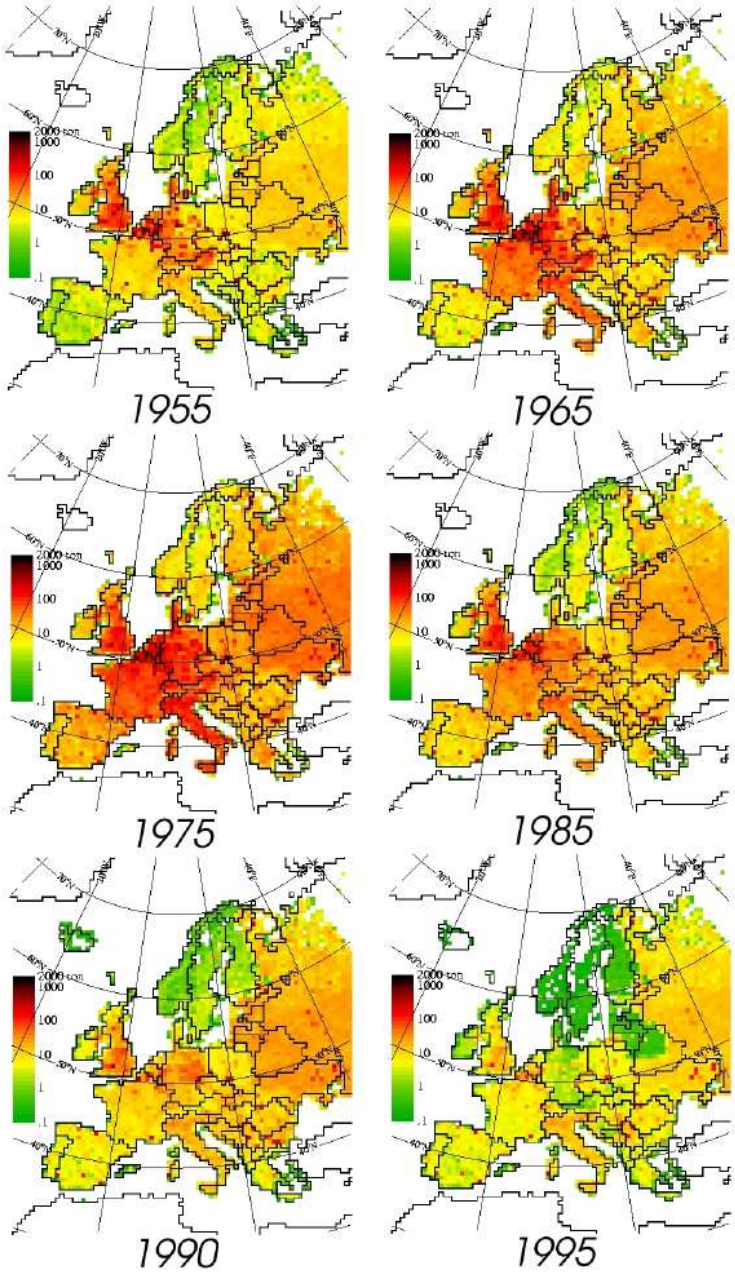

Figure 3: Estimated annual atmospheric lead emissions for the model grid cells (data from Pacyna and Pacyna, 2000).

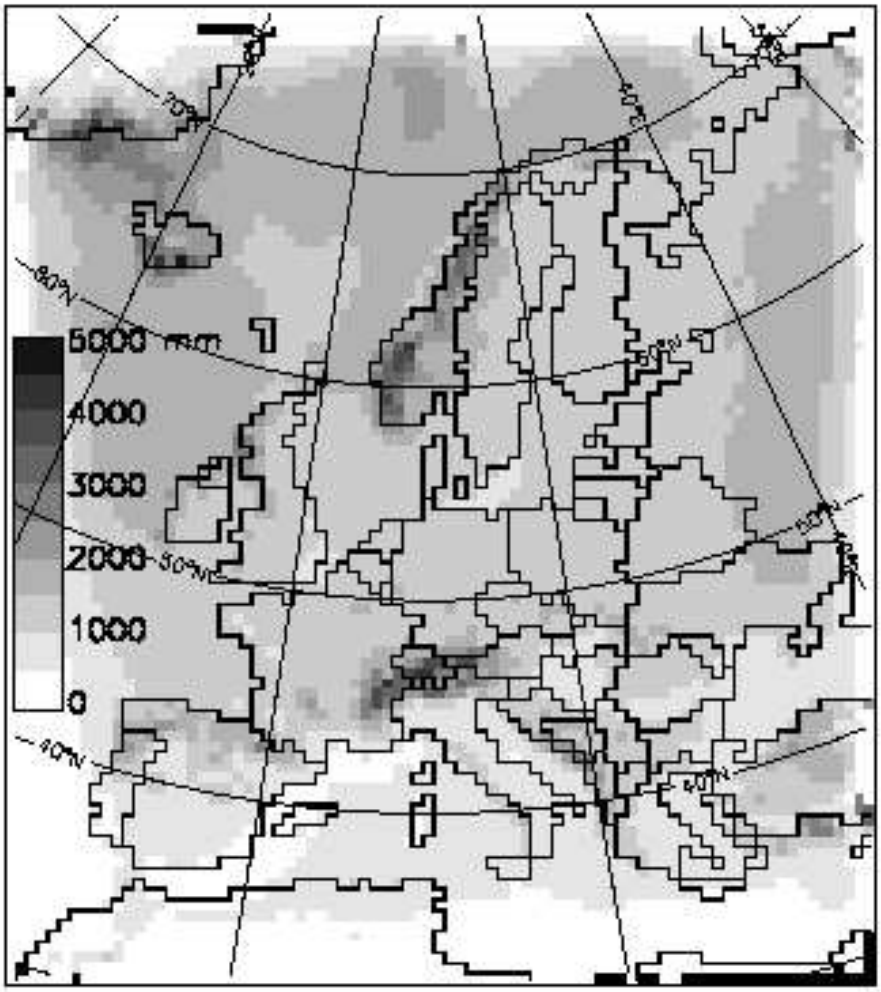

Figure 4: Average simulated precipitation in 1958-1995.

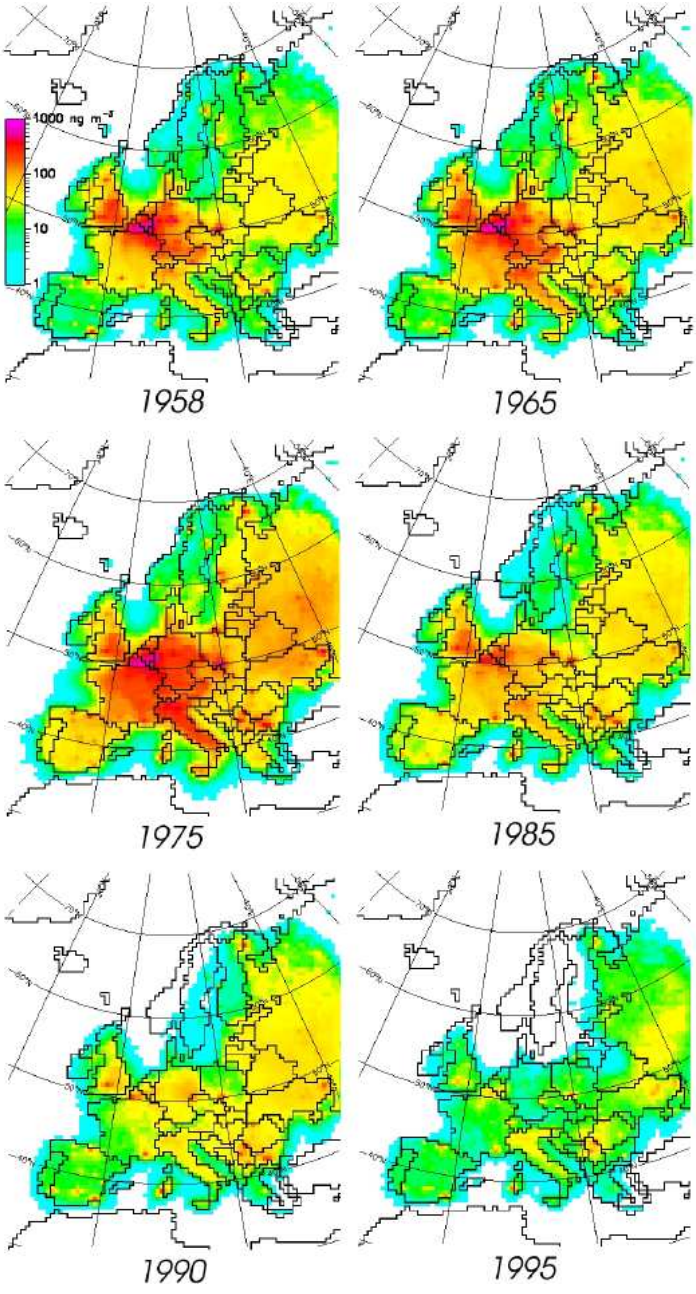

Figure 5: Mean annual simulated air lead concentration

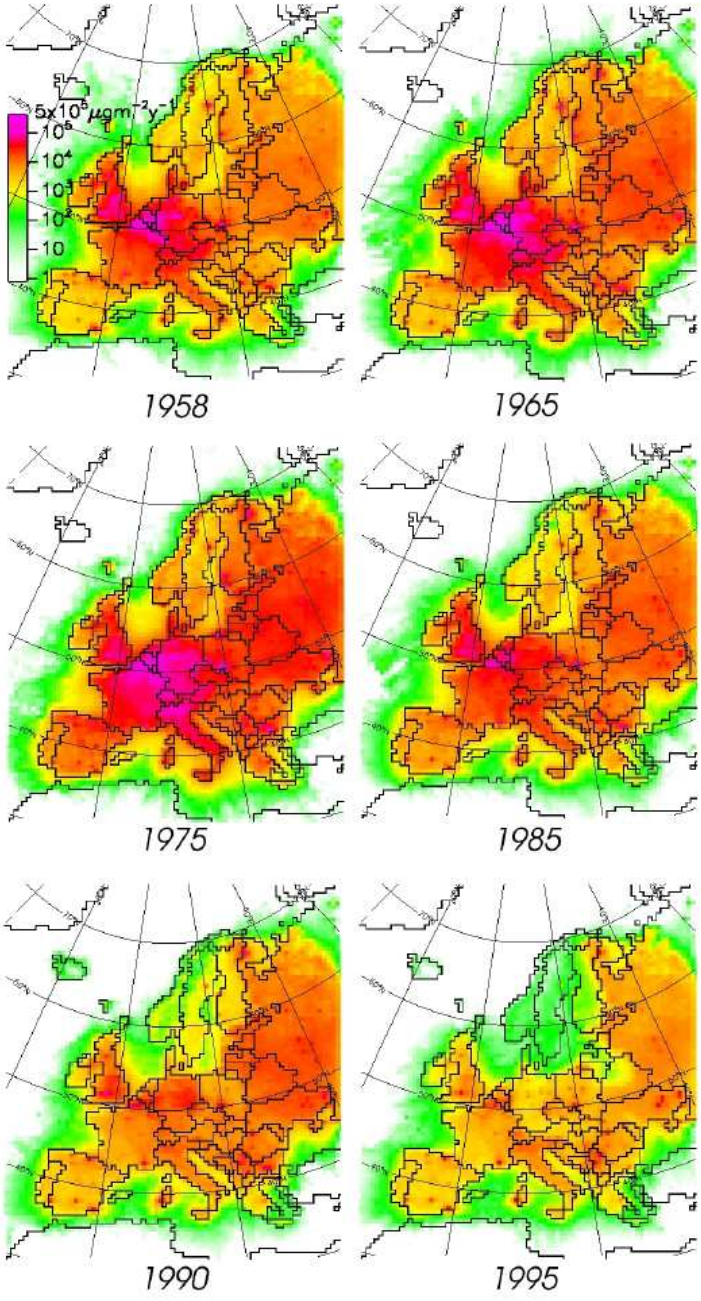

Figure 6: Mean annual simulated air lead deposition rates

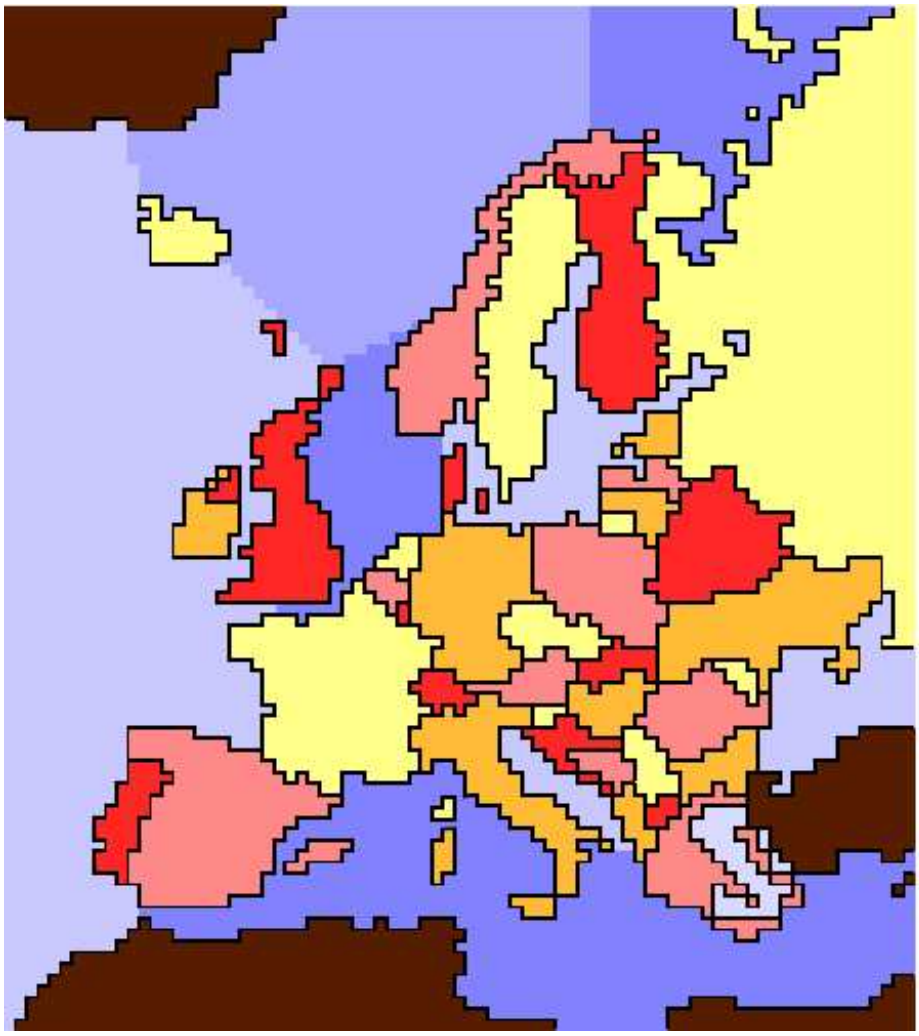

Figure 7: Countries and regions for which source-receptor relations were calculated.

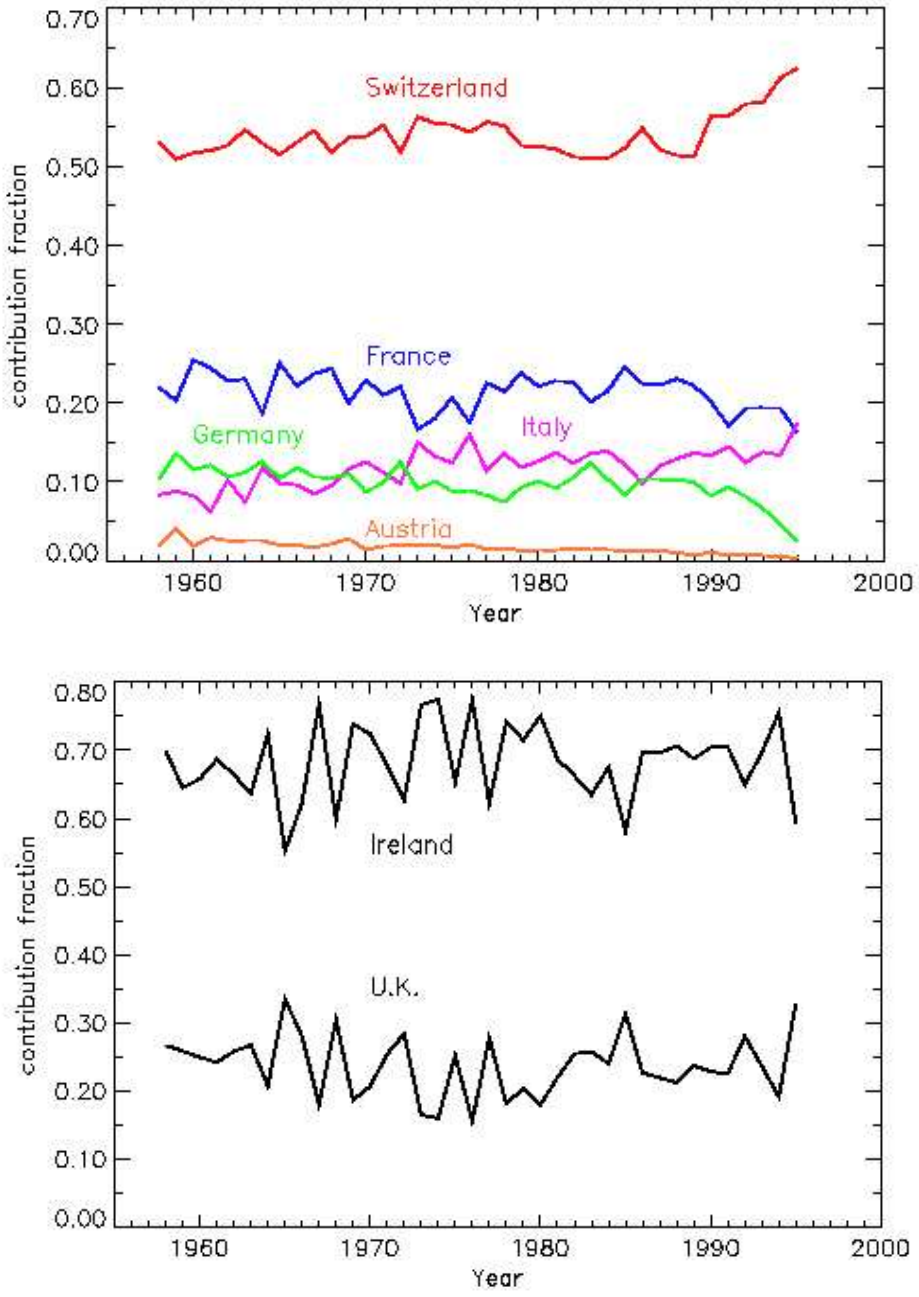

Figure 8: Annual fraction of source-country contribution to lead deposition in (a) Switzerland (top) and (b) Ireland (bottom).

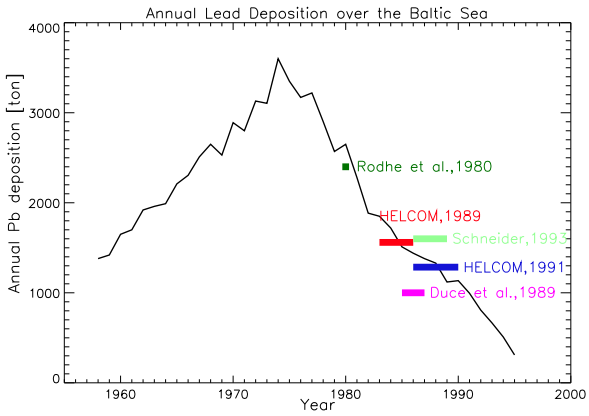

Figure 9: Annual lead deposition over the Baltic Sea, from measurement-based estimates (colored bars) and our simulations (line).

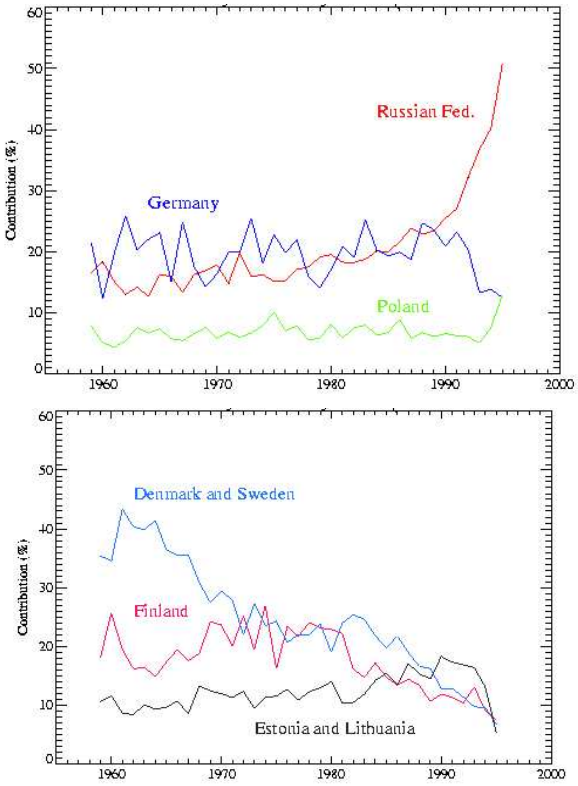

Figure 10: Fraction contributed by each country to lead deposited in the Baltic Sea

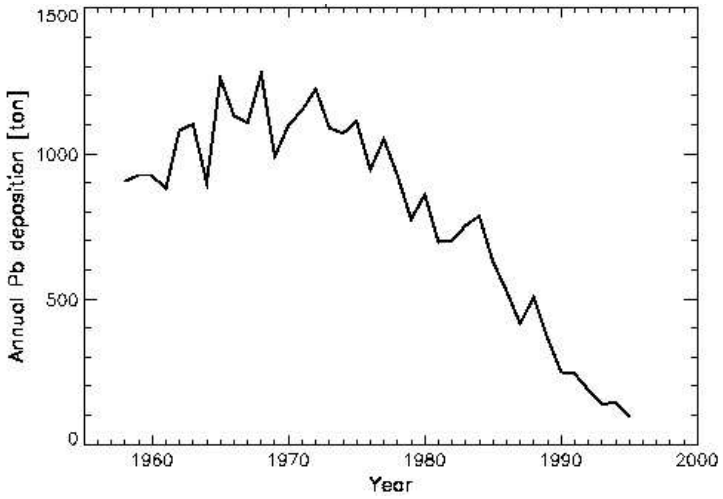

Figure 11: Annual simulated lead deposition over the North Sea.

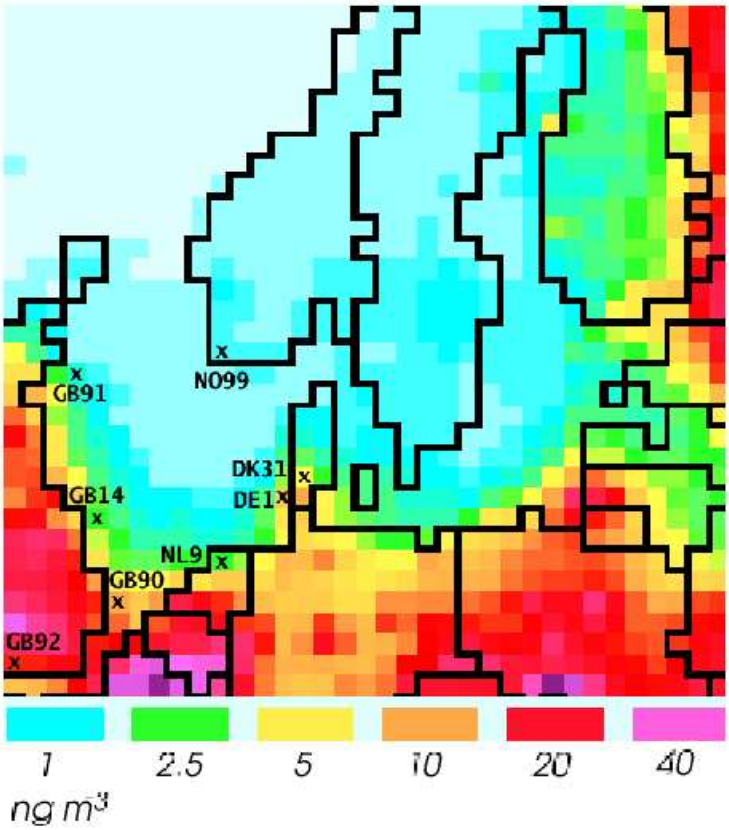

Figure 12: Mean simulated air concentration in 1995 and location of EMEP measurement stations in the North Sea and Baltic Sea regions.

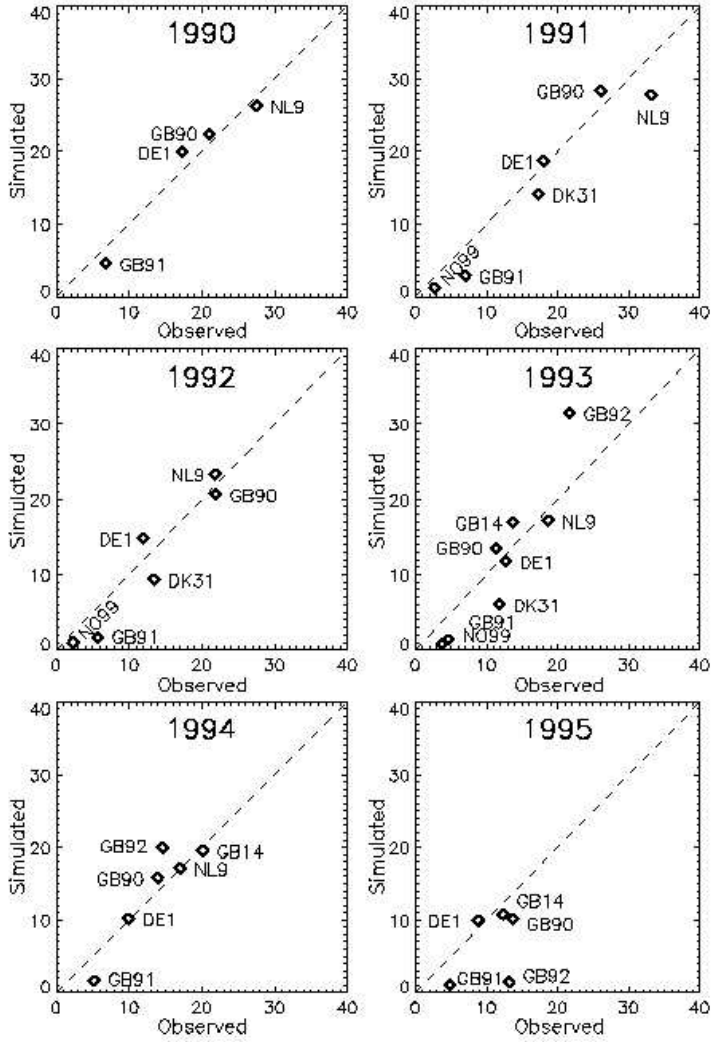

Figure 13: Observed and simulated annual mean lead air concentrations at EMEP stations at coastal locations of the North Sea and English Channel.

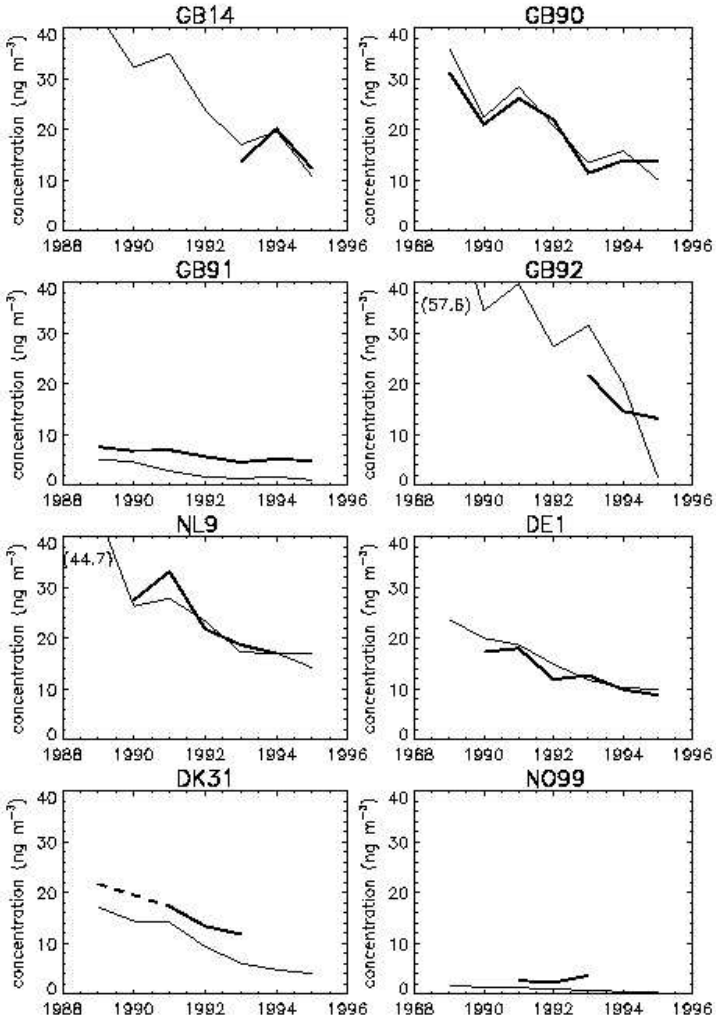

Figure 14: Observed (bold line) and simulated (thin line) lead air concentrations (ng/m3) at EMEP stations on the North Sea coast and vicinity. All yy axis cover the same range, to allow for better comparison between plots. The dashed line for station DK31 connects two data points between which one data point is missing (1990).

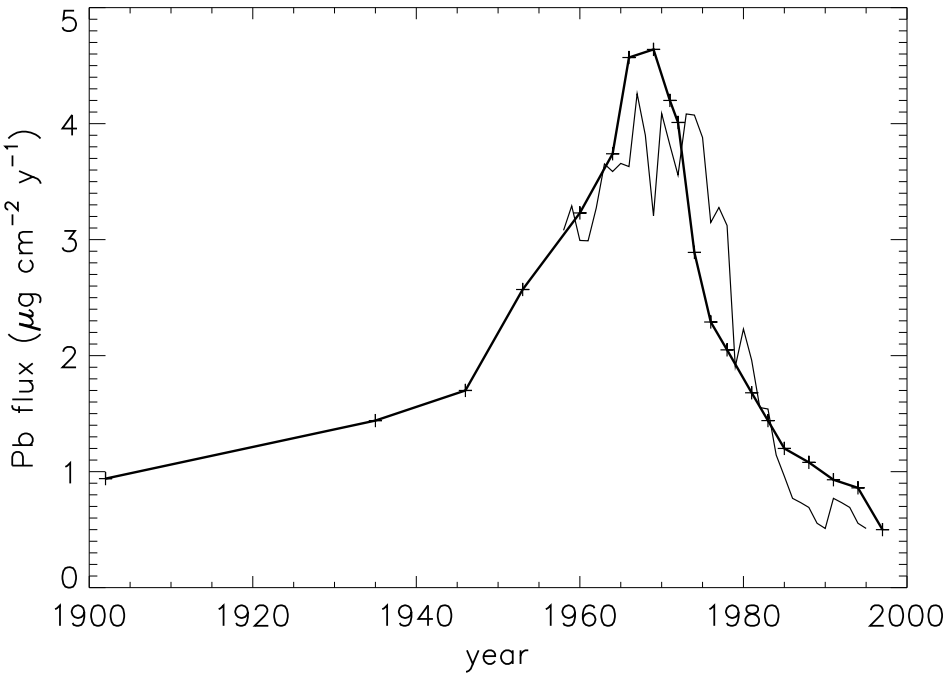

Figure 15: Measurement-based (bold line) (data from Goodsite et al., 2001) and simulated (thin line) deposition rate at Storelungmose, Denmark.

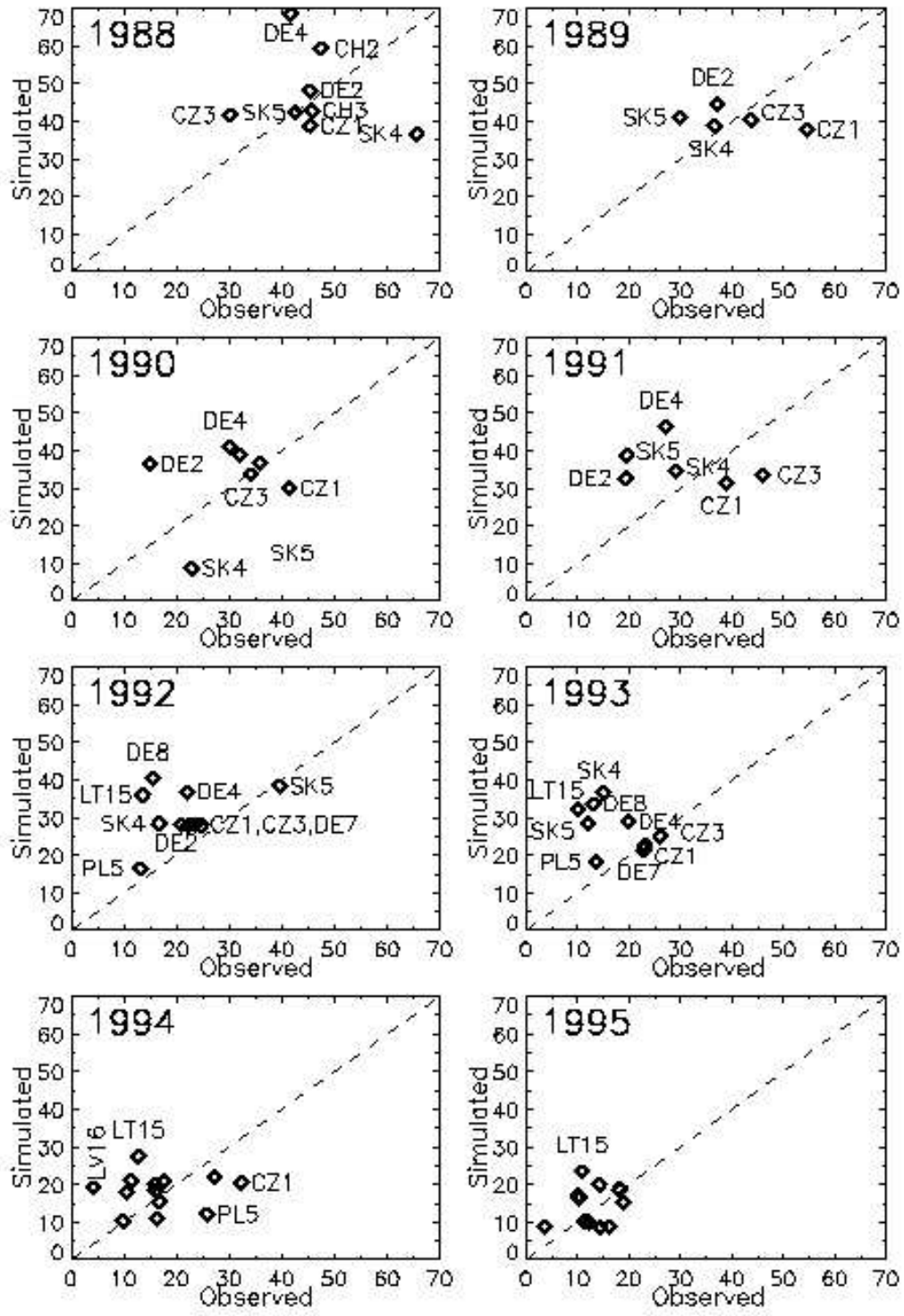

Figure 16: Observed and simulated lead air concentrations (ng/m3) at EMEP stations at various Center and Center-East European locations.

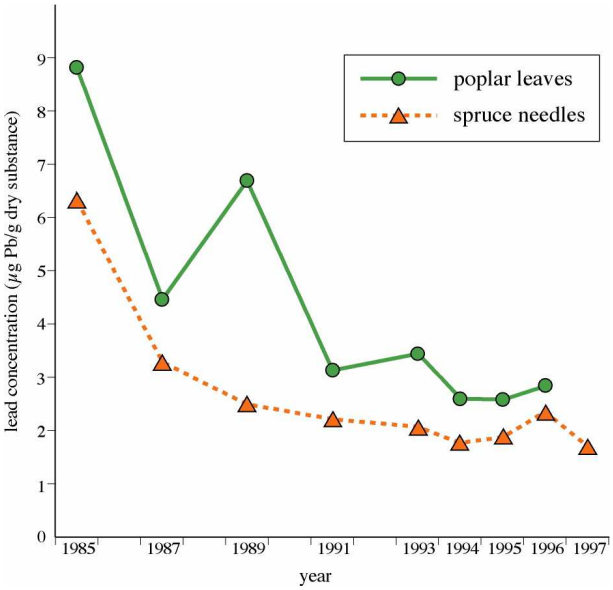

Figure 17: Lead concentration in spruce (*Picea abies*) sprouts and poplar (*Populus nigra*) leaves in urban areas in Saarland/germany. Data source: Umweltprobenbank 1999.

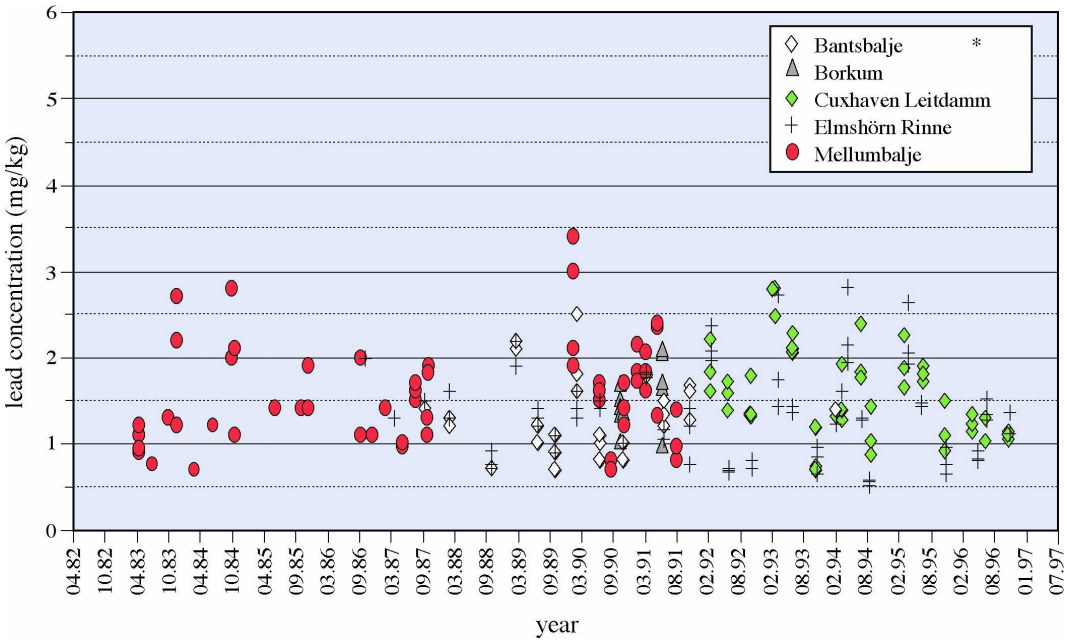

Figure 18: Lead Concentration in Blue Mussels (*Mytilus edulis*) in the North Sea. Data source: Ministry of Ecology of Niedersachsen 1999.

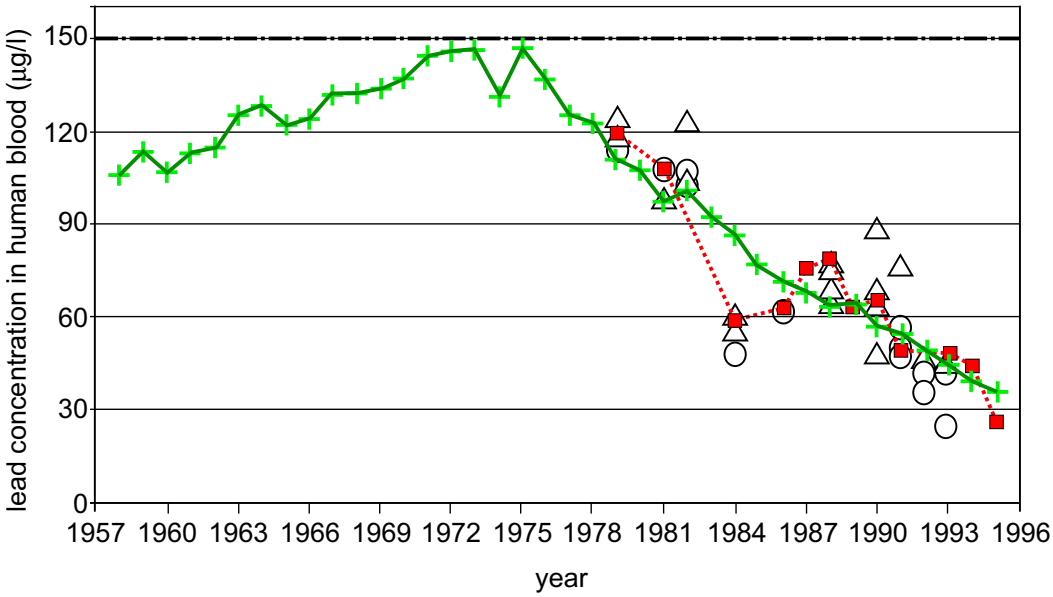

Figure 19: Blood lead levels measured in the student population of the University of Münster, Germany (red dashed line); and in different measurement campaigns in Germany (adults: triangles; children: open circles), from various studies in Germany. Data source: Heinzow 1998. Also shown is an estimate of past blood lead concentrations in adults based on the Münster measured data, assuming a linear relation between mean lead concentrations in blood and modelled mean lead concentrations in the atmosphere on a given year in the model grid cell where Münster is located (green line).

Human Blood Lead Level in Germany. Data source: Human-Probenbank Münster 2001

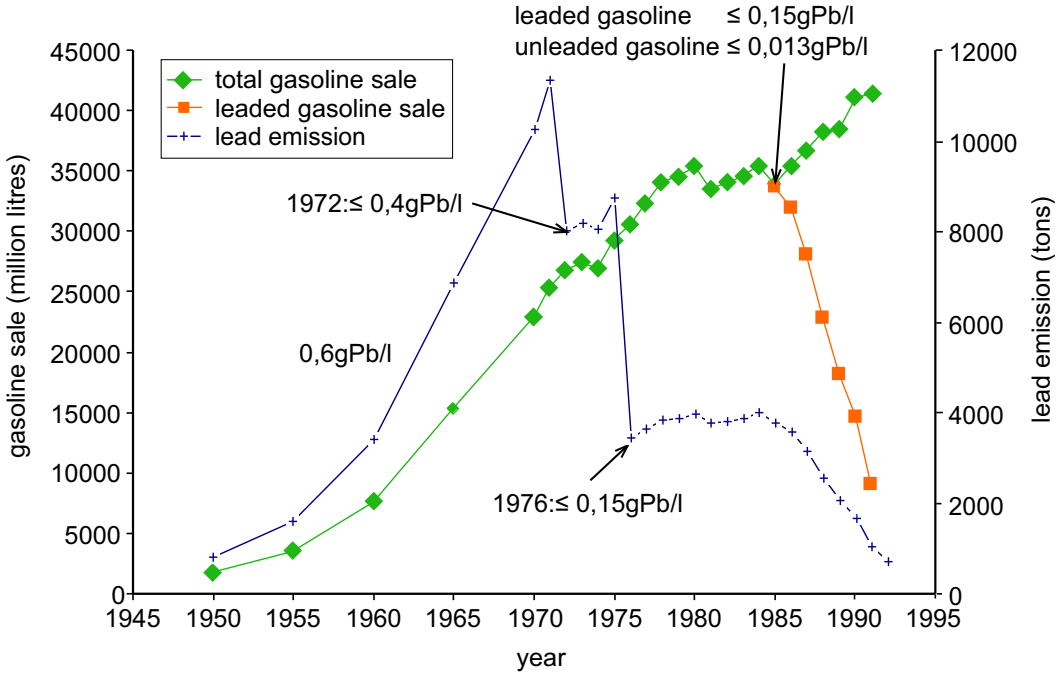

Figure 20: Gasoline Sales and Lead Emissions in Germany. Data source: Mineralölwirtschaftsverband 2000.