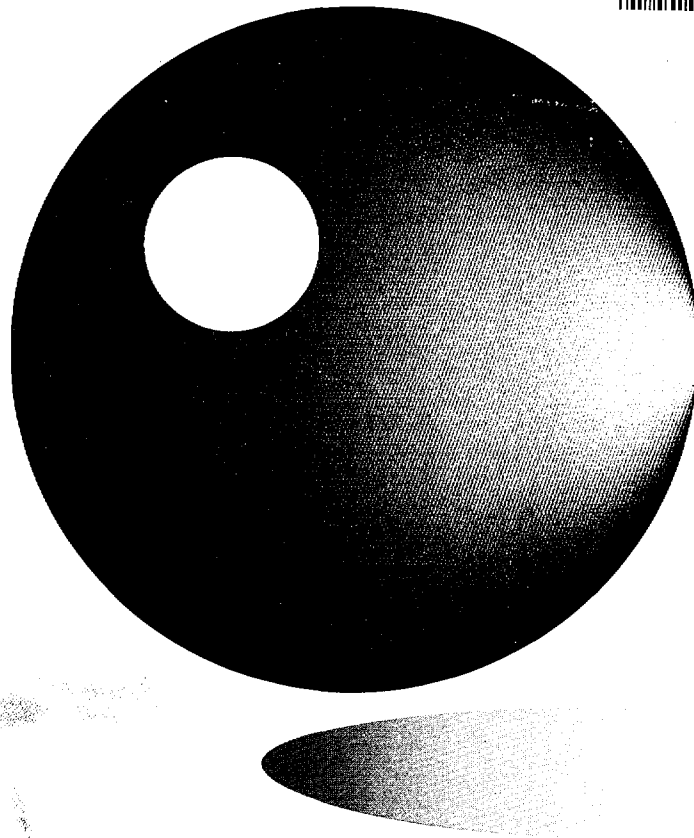




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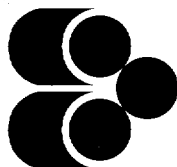
**Lead isotopes as an environmental
indicator of pollution sources**

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Abstract <p>An interesting question when determining and quantifying pollution is the characterisation of the sources. Also, is the pollution locally derived or is it longtransported? Analyses of Pb isotope ratios and Pb concentrations on airfilter from Oslo, Norway, together with analyses of coal of different origin and wood, show that the decrease in total Pb in the first half of the 1990's is not due only to the reduction of leaded petrol but that there are other important sources for the pollution which have been cut down. The reduction in emissions from coal burning in Europe can be seen on airfilter from Oslo and also the following reduction in leaded petrol. An isotopic study of lead on airfilter from the countryside outside Oslo implies that the finer particles have been atmospherically long transported while the coarser particles are of a more local origin, e.g. from domestic burning of wood. The above results show that the use of a natural isotopic system like Pb is a powerful tool for e.g. the determination of pollution, routes of its transportation, for the monitoring of processes of the present, and for provenance determinations.</p>			
Key Words Lead isotopes, pollution sources, atmospheric transport			
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Lead isotopes as an environmental indicator of pollution sources

NFR project no.: 110727/730

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1 Background

Lead is a dangerous factor for pollution of the environment, but has the advantage that it can be used as a tracer in environmental studies by its naturally occurring isotopic system. Despite a decrease in the emission of different pollutants in the western hemisphere we may in the coming years be exposed to an increase in the global transport of e.g. heavy metals. Today there are strict laws regulating the handling of waste and emission in the developed countries, but not in the former Eastern European and the developing countries.

Lead has been utilised by mankind for thousands of years. Some of the applications have been water-pipes, kitchenware, paint, for medical purposes and as a sweetening agent. Lead is, however, highly poisonous and thus having drastic effects when reaching high concentrations in the body. In the 20th century, learning the effects of lead led to a withdrawal of its use in most applications. In one area, however, lead experienced a renaissance: as an anti-knock additive in petrol. As such, it was first introduced in the 1920:ies and reached its peak around 1970 when lead free gasoline was introduced. In 1972 world production of lead was about 3.75 million tons of which about 10% was used for lead alkyl additives (Chow et al. 1975).

The emission of lead from cars together with the emission from the industry, smelters and fly-ash taken together in the 1970-80:s contributed to a deposition of lead in the environment of a much greater magnitude than ever before in history. Moreover, the change from centrally planned to market oriented economy in the former Eastern European countries and their need for energy have in some cases increased the use of coal which contain lead. Long neglected waste

dumps are slowly leaking, polluting water and environment with lead and other heavy metals. A study from North America shows that the burning of coal releases lead concentrations as high as 150 ppm in fly ash and 360 ppm in soot near coal-burning electric power plants (Chow & Earl, 1972).

Apart from being deposited near the source of emission, lead may be transported over wide areas. Studies by Sturges & Barrie (1989 a, b) showed that it was possible to distinguish between lead from Canadian car exhaust, Canadian smelters and eastern American sources. Moreover, in the Arctic they found great differences between lead from two widely separated Canadian Arctic stations, from a station in Spitsbergen and one in northern Norway. New threats in these aspects are the industrialisation of Asia where the emission from coal burning in China and India will contribute to an enormous global contamination.

2 The geochemistry of lead

The element lead is occurring naturally with four Pb-isotopes with the following distribution; ^{204}Pb 1.4%, ^{206}Pb 24.1%, ^{207}Pb 22.1% and ^{208}Pb 52.4%. The ratio between the different lead isotopes varies in different geological systems due to the formation of ^{206}Pb and ^{207}Pb when ^{238}U and ^{235}U , respectively, decays while ^{208}Pb is a product of the radioactive decay of ^{232}Th . ^{204}Pb is the only isotope of lead which gets no addition from radioactive decay and thus can be looked upon as stable. Because of its natural variations in isotopic ratios, lead can be used as a tracer in environmental studies.

3 Project objectives

An interesting question when determining and quantifying pollution is the characterisation of the sources. Also, is the pollution locally derived or is it longtransported? The overall goal of this project was to develop and apply radioisotope ratios as a technique to study past and present changes of air pollution in the North. The principal method used in this project is the analysis of isotopes and concentration of lead in air-filters and coal in order to try to differentiate between pollution originating from west (England) and from Southeast (Central Europe). The lead isotope signature differ significantly between lead additives to gasoline, coals, lead bearing ores etc.

4 Analytical procedures

The air filters were leached with 10 ml 10% HNO₃ by shaking overnight using a mechanical shaker. 5 ml of the solution was pipetted out and evaporated to dryness. For coal, ash and wood samples the organic material was removed with a low temperature plasma asher. The residue was treated with concentrated HNO₃ which was then evaporated to dryness.

In order to avoid disturbances during the determination of concentration and isotopic composition, the lead had to be separated from most other elements present in the sample. For this purpose a chemical technique called ion extraction chromatography was applied. The residues from the ashing or extraction procedure were dissolved in 3 M HNO₃ and loaded onto columns packed with a special resin which retained Pb and let most other elements pass unretained through. After rinsing out the remaining unwanted other elements, the lead was washed out of the column with diluted ammonium carbonate solution. The collected Pb-fraction was then evaporated to dryness.

The lead concentration was determined by flameless atomic absorption spectrometry (FAAS) and the isotopic composition by thermal ionisation mass spectrometry (TIMS). The precision of the FAAS analysis is considered to be within $\pm 5\%$ and the TIMS analysis to be within $\pm 0.01\%$.

5 Results from the investigation

5.1 Comparison of Pb on airfilters from the countryside and Oslo town

In a study of Pb on airfilters from the town of Oslo and from the countryside we have tried to discriminate between long-transported and local pollutants, and to see if there is a variation over the year.

The size and type of particles have implications for the Pb isotope ratio and the Pb concentration. In a first test we compared coarse and fine particle filters from a Dichotomous sampler with a cut-off at 10 μm on samples from 1992-93. Inside Oslo town (Kirkeveien) the filters have similar ²⁰⁶Pb/²⁰⁴Pb ratios, 17.182 for the fine and 17.201 for the coarse particles (Figure 1).

The Pb concentrations are however very different, 5.27 $\mu\text{g/g}$ for the fine and 2.97 $\mu\text{g/g}$ for the coarse particles (Table 1). In the filters from the countryside (Gardermoen) there is a marked difference between the Pb ratios. The fine particles have a ²⁰⁶Pb/²⁰⁴Pb ratio of 16.965 while

the coarse particles have a value of 17.158. The filter with fine particles have moreover 3 times higher concentration of lead (0.16 $\mu\text{g/g Pb}$) than the coarse one (0.06 $\mu\text{g/g Pb}$).

One source is obviously dominating in Oslo town, as seen from the similar Pb isotope ratios on both the coarse and the fine filters. At the countryside with less pollution in the air, both filters have lower Pb concentrations and the town source will thus be of less importance. The Pb concentration is also about 50 times lower for the coarse particles in the countryside compared to the concentration of the town filter.

The lower $^{206}\text{Pb}/^{204}\text{Pb}$ ratio for the fine filter from the countryside reveals the presence of another source. The finer particles deposited on this filter are probably long transported and can be distinguished here because of the low background from the town area which in the town with its high level of pollution is masking this foreign source. The low Pb ratio on the fine filter from Gardermoen may reflect the impact of leaded petrol.

The dominating source in Oslo town with $^{206}\text{Pb}/^{204}\text{Pb}$ ratios about 17.2, or higher, is most probably of local origin. Leaded petrol has a quite low $^{206}\text{Pb}/^{204}\text{Pb}$ value of about 16.0 (Precambrian Pb from Broken Hill, Australia, e.g. Faure 1986) and is probably indicated on the Gardermoen fine filter, partly from long transport. On the Oslo fine filter this imprint from traffic is probably seen in a small deviation from the dominating value of the coarse filter (from a $^{206}\text{Pb}/^{204}\text{Pb}$ value of 17.201 to 17.158, Table 1).

5.2 Pb on airfilters from two sites with heavy traffic in Oslo town

Air filters collected at two sites with high traffic intensity in the town of Oslo show a constant decrease in Pb concentration from 1992 to 1995 (Figure 2). These data are in accordance with the conventional wisdom about the effect of change from leaded to unleaded petrol. However, analyses of the $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratio on same air filters from Kirkeveien and Gamlebyen in Oslo show a decrease from 1992 to 1993 and then an increase in 1994. In 1995 exceeding the values from 1992 (Figure 3).

The strong decrease in Pb concentration from 1992 to 1993 (Figure 2) indicates that one major source is losing its impact. This decrease is coupled to a decrease in the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio from 17.19 to 16.81 (Figure 3). However, further decrease in Pb concentration during 1994 and 1995 is matched by an increase in $^{206}\text{Pb}/^{204}\text{Pb}$ ratio. A decrease in coal burning in Central Europe during 1992 to 1995 would lower the emission of Pb transported to Oslo. In this case decreased use of coal in the former Eastern European countries results in decreasing Pb

concentration from 1992 and onwards. Less European coal with a $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratio of about 18.4 (Table 1), would result in a tilt of the total $^{206}\text{Pb}/^{204}\text{Pb}$ ratios towards that of leaded petrol. The Pb isotope ratios will thus decrease towards lower petrol values but the turning over to more Pb free petrol will have the effect of increasing the Pb ratios while the decrease in Pb concentration continues. As the Pb concentration is quite low from 1994 other small sources may be seen now. Like e.g. the burning of wood, with a higher $^{206}\text{Pb}/^{204}\text{Pb}$ ratio of about 18, for domestic heating.

Indeed, in some areas, such as Scandinavia, burning of wood can be an important source of atmospheric lead. An emission factor of 4.7 g Pb/tonne of wood burnt was suggested for fireplaces already in the 1980's (e.g. Pacyna, 1986). Further studies concluded that this factor may range from 1.0 to 5.0 g Pb/tonne of wood burnt (Nriagu and Pacyna, 1988) depending on parameters related to the content of Pb in wood and combustion conditions. The emission factor for Pb during combustion of wood in fireplaces are comparable with the Pb factors for combustion of both coal (1.0 - 10.0 g/tonne) and oil (2.0 - 6.0 g/tonne) as described by Nriagu and Pacyna (1988). Taking into account the rather large amount of wood burnt in Scandinavia, particularly in winter, this source can be an important contribution to the total budget of atmospheric lead in the region.

5.3 Compilation of Pb results from various sources

A plot of the $^{208}\text{Pb}/^{206}\text{Pb}$ ratio versus the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio from various sources of Pb (Figures 4 & 5) show that wood (tree trunk) is closer to modern Pb (from soil) and coal Pb while the Pb ratio in the bark seems to be more affected by atmospherically transported petrol Pb. A similar pattern is valid for the air filters; the filter with finer particles from the countryside is closer to an air transported Pb petrol value, while the coarse filter from the countryside and both filters from Oslo town have more of a bark/wood signature. The Pb concentration in the bark is about 10 times higher compared to that in the trunkwood. Contamination in Oslo by longtransported Pb from the burning of coal, in England or Poland and Czechia, may have had some influence. Maybe not so much today but in earlier years when coal was used to a larger extent. Long-time deposition of this pollution and uptake by biologic matter may release this memory effect when burning wood. Most English coal mines were closed during the 1980:ies and are now mainly causing local damage due to acid mine drainage due to neglected maintenance of the mines.

It should also be added that a progress has been made in the Eastern European countries during the last 5 years with respect to the improvement of emission control effectiveness. Major point sources, such as electric- and cogeneration power plants, smelters, cement plants, etc. are now

equipped with at least electrostatic precipitators with high efficiency for removal of dust containing Pb, among other pollutants.

Soot from domestic heating, by burning wood, was collected in two houses in the Oslo-area. In one case the wood originated from the Oslo community area while in the other case it was brought in from the countryside in Telemark, southern Norway. There is a big difference in Pb concentration between the two samples. While the Oslo soot has a concentration of about 64 ppm Pb, the Telemark soot sample carries only about 14 ppm Pb (Table 1). There are of course different parameters influencing these results like e.g. the efficiency of the furnace but the gap between the results is significant.

The $^{206}\text{Pb}/^{204}\text{Pb}$ ratio of the Oslo soot has the lower value of the two (Table 1) thus indicating an affinity to leaded petrol from traffic while the Telemark soot has a higher $^{206}\text{Pb}/^{204}\text{Pb}$ ratio more towards that of European coal. In Figure 5 these relations are clearly shown.

5.4 Pb on airfilters from Gardermoen

Pb was also analysed on air filters collected at Gardermoen between 1958 and 1982. The filters were originally sampled by FFI at a daily basis. For the investigation we selected 2 filters a year, from the middle of April and the middle of August, yearly in 1958 to 1960 and 1980 to 1982, and every fifth year between 1960 and 1980 (Table 2).

A plot of the Pb concentration vs. time (Figure 6) shows a minima in the middle of the 1960:ies but then there is an increase up to the early 1980:ies when there is a sharp drop where we got the lowest analysed concentration values. A comparison with Figure 7, showing the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio vs. time for the air filters, may give a small indication of an increase in the Pb ratios. Between 1965 and 1982 there was a strong increase in the consumption of petrol, and during this time interval the petrol was leaded. The isotopic composition of leaded petrol would on the other hand tilt the analysed Pb ratios towards lower values.

In Figure 7 there is a possible minima in the middle of the 1960:ies just like in Figure 6. The question is if these changes are due to a change, on local or on regional scale. The number of Pb analyses from the Gardermoen locality are still very sparse and heavier traffic with petrol engined aircrafts during a day or two may have a sudden impact on the analytical values. This influence is a fact which has to be checked out more carefully before any more samples from between 1958 and 1982 are analysed from Gardermoen.

On Figure 8, $^{206}\text{Pb}/^{204}\text{Pb}$ ratio vs. Pb concentration, the main part of the filters have a Pb concentration of less than $20\ \mu\text{g/g}$ and the main part of the filters a $^{206}\text{Pb}/^{204}\text{Pb}$ ratio between 17.3 and 18.0. In comparison air filters from Oslo and Gardermoen between 1992 and 1995 have $^{206}\text{Pb}/^{204}\text{Pb}$ ratios between 17.2 and 16.8 (Figs. 1 & 3) which is substantially lower than 10 to 20 years back. The Pb concentration in 1993 was 0.06 and $1.16\ \mu\text{g/g}$ for two filters from Gardermoen (Fig. 1 & Table 2) to be compared with the up to $20\ \mu\text{g/g}$ for 1958 to 1982. A reservation has to be made here for the high values as they may mirror input from petrol engined aircrafts. The most interesting period between 1982 and 1992 has not been investigated yet since there is still a lack of air filters for this period.

6 Summary

Lead isotope ratios can be an environmental indicator for:

- the origin of toxic air pollutants transported to Norway from other countries, and
- historical changes of the magnitude of air contamination by lead in the country due to local and international environmental policies.

The use of radioisotopic ratios as a technique developed within the present project can be applied to study any impact of emission reductions for toxic compounds in Europe on their distribution and levels in the North. This is particularly important in a view of preparations of the UNECE protocols on heavy metal emission reductions.

7 Acknowledgements

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Table 1. Pb isotope ratios of airfilters, coal, flyash, bottomash and soot.

Sample	Date	206/204	207/204	208/204	206/207	$\mu\text{g/g}$	208/206
Kirkeveien	26-27/11-92	17.182	15.509	36.831	.1.108	5.27	2.144
Fine, Nov-92							
Kirkeveien	26-27/11-92	17.201	15.506	36.838	1.109	2.97	2.142
Coarse, Nov-92							
Gardermoen	28-29/9-93	16.965	15.478	36.606	1.096	0.16	2.158
Fine, Sep-93							
Gardermoen	28-29/9-93	17.158	15.512	36.822	1.106	0.06	2.146
Coarse, Sep-93							
Blindfilter							
Mars -96						(1.8 ng)	
Coal - COUS	8/2-96	18.458	15.595	38.363	1.184	11.7	2.078
Bott.ash - BAUS	8/2-96	18.342	15.577	38.222	1.177	1.72	2.084
Fly ash - FAUS	8/2-96	18.555	15.503	38.388	1.189	4.33	2.069
Coal Halemba	Sept. 96	18.533	15.602	38.398	1.189	7.20	2.072
Flyash Halemba	Sept. 96	18.572	15.605	38.456	1.191	5.60	2.071
Coal Laziska	Sept. 96	18.519	15.603	38.362	1.188	21.8	2.071
Flyash Laziska	Sept. 96	18.486	15.618	38.406	1.185	9.80	2.078
Coal Chorzow	Sept. 96	18.464	15.623	38.384	1.183	44.2	2.079
Flyash Chorzow	Sept. 96	18.489	15.623	38.428	1.184	19.8	2.078
Coal Lagisza	Sept. 96	18.531	15.627	38.422	1.187	13.4	2.073
Flyash Lagisza	Sept. 96	18.566	15.618	38.435	1.190	18.0	2.070
Coal C6		18.542	15.590	38.254	1.189	0.44	2.063
Coal A24		19.437	15.645	38.361	1.242	0.66	1.974
C 5.1		18.457	15.637	38.157	1.180	21.2	2.067
C 8.2		17.912	15.569	37.750	1.150	13.9	2.108
C 10.2		18.391	15.596	38.342	1.179	0.92	2.085
C 11.1		18.287	15.567	38.193	1.175	2.62	2.089
C 11.2		18.385	15.597	38.318	1.179	22.5	2.084
C 13.1		18.181	15.532	38.088	1.171	82.4	2.095
Broken Hill	Litt. value	16.002	15.389	35.657	1.040		2.228
Stacey-Kramer	Litt. value	18.700	15.628	38.630	1.197		2.066
Common Pb							

Table 1. Pb isotope ratios of airfilters, coal, flyash, bottomash and soot.

	Dato	206/204	207/204	208/204	206/207	Pb ppm	208/206
FP-DIC/fin fraksj							
Kirkeveien	20-21/11-92	17.155	15.536	36.883	1.105	4.16	2.150
Kirkeveien	29-30/11-92	17.194	15.552	36.954	1.107	5.71	2.149
Kirkeveien	29-30/12-92	17.084	15.511	36.776	1.102	11.3	2.153
Kirkeveien	29-30/3-93	16.880	15.497	36.565	1.090	2.65	2.166
Kirkeveien	31/3-1/4-93	16.894	15.519	36.640	1.090	3.81	2.169
Kirkeveien	9-10/12-93	16.814	15.523	36.581	1.084	5.35	2.176
Kirkeveien	21-22/12-93	16.832	15.528	36.604	1.085	5.49	2.175
Kirkeveien	7-8/3-94	16.919	15.523	36.661	1.091	2.12	2.167
Kirkeveien	24-25/3-94	16.858	15.498	36.551	1.089	1.73	2.168
Kirkeveien	28-29/10-94	17.209	15.532	36.937	1.109	0.62	2.146
Kirkeveien	24-25/11-94	17.186	15.526	36.901	1.108	0.44	2.147
Kirkeveien	30-31/1-95	17.264	15.533	36.982	1.112	1.59	2.142
Kirkeveien	26-27/2-95	17.228	15.556	37.009	1.108	0.58	2.148
Gamlebyen	21-22/12-92	17.132	15.551	36.913	1.103	5.88	2.155
Gamlebyen	27-28/12-92	17.090	15.525	36.806	1.102	6.86	2.154
Gamlebyen	29-30/12-92	17.088	15.554	36.878	1.100	8.89	2.158
Gamlebyen	30-31/12-92	17.103	15.527	36.825	1.103	3.72	2.153
Gamlebyen	5-6/3-93	16.899	15.504	36.595	1.091	2.70	2.166
Gamlebyen	29-30/3-93	16.914	15.511	36.630	1.091	2.04	2.166
Gamlebyen	31/3-1/4-93	17.008	15.532	36.770	1.096	1.86	2.162
Gamlebyen	9-10/12-93	16.873	15.517	36.620	1.088	2.21	2.170
Gamlebyen	21-22/12-93	16.877	15.503	36.576	1.090	0.62	2.167
Gamlebyen	16-17/2-94	17.088	15.518	36.774	1.102	2.35	2.152
Gamlebyen	17-18/3-94	16.971	15.543	36.759	1.093	1.24	2.166
Gamlebyen	24-25/3-94	16.871	15.501	36.570	1.089	0.62	2.168
Gamlebyen	24-25/4-94	17.125	15.520	36.865	1.104	1.11	2.153
Czech bl. coal	Okt-96	18.406	15.627	38.428	1.178	20.5	2.088
Czech br. coal	Okt-96	18.388	15.551	38.282	1.183	0.16	2.082
Svalb. SV 22	Okt-96	18.084	15.458	37.962	1.170	0.30	2.099
Svalb. SV 26	Okt-96	18.103	15.438	38.122	1.173	0.66	2.106
Svalb. SV 27	Okt-96	18.038	15.457	37.867	1.167	0.12	2.099
Bjørg soot	Wood Oslo	17.978	15.586	37.821	1.153	64.4	2.104
Harald soot	Wood Telemark	18.178	15.575	37.898	1.167	14.4	2.085

Table 2. Pb isotope ratios of airfilters from Gardermoen, NW of Oslo.

Sample	Dato	206/204	207/204	208/204	206/207	Pb ppm	208/206
G 292 L	7/4-58	16.763	15.454	36.484	1.085	17	2.176
G 419 L	11-12/8-58	17.597	15.550	37.458	1.132	7.3	2.129
G 1149 L	11-12/8-60	17.765	15.558	37.626	1.142	5.9	2.118
G 2829	6-7/4-65	17.317	15.517	37.100	1.116	81.4	2.142
G 2956	11-12/8-65	17.445	15.518	37.222	1.124	3.5	2.134
G 4656	10-11/4-70	17.688	15.566	37.561	1.136	5.4	2.124
G 4779	11-12/8-70	17.241	15.508	37.012	1.112	12.4	2.147
G 6116	10-11/4-74	17.795	15.570	37.539	1.143	19.3	2.110
G 6243	15-16/8-74	17.663	15.553	37.454	1.136	16	2.120
G 8308	10-11/4-80	17.452	15.553	37.210	1.122	1.4	2.132
G 8435	15-16/8-80	17.530	15.538	37.183	1.128	14.3	2.121
G 8673	10-11/4-81	17.718	15.567	37.385	1.138	37.4	2.110
G 8800	15-16/8-81	18.005	15.584	37.778	1.155	1.4	2.098
G 9038	10-11/4-82	17.773	15.564	37.543	1.142	1.1	2.112

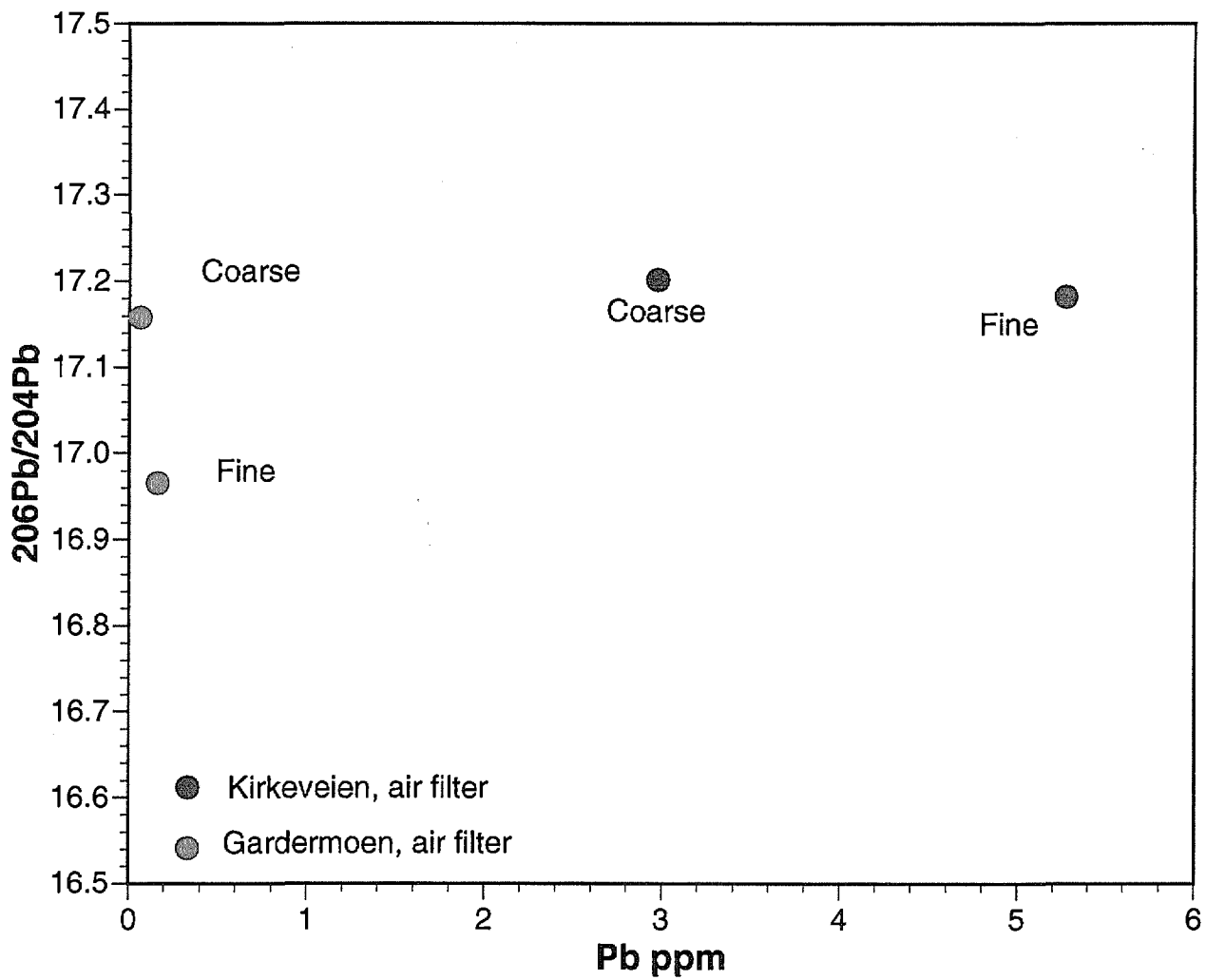


Figure 1. Pb isotope ratios vs. Pb concentration for coarse and fine particles sampled on air filters in Oslo town and from the countryside.

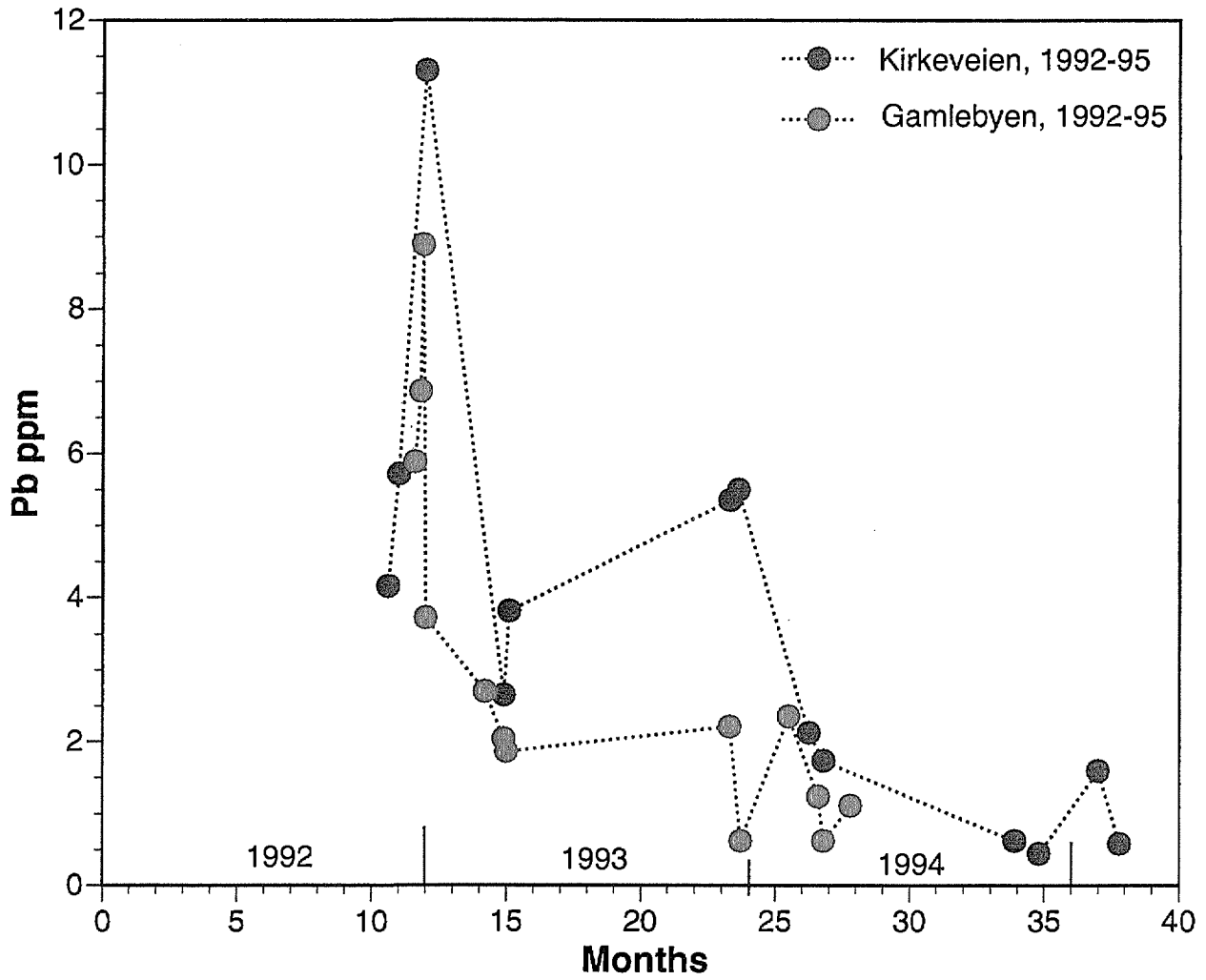


Figure 2. Pb concentration for air filters collected at two sites with heavy traffic in Oslo town.

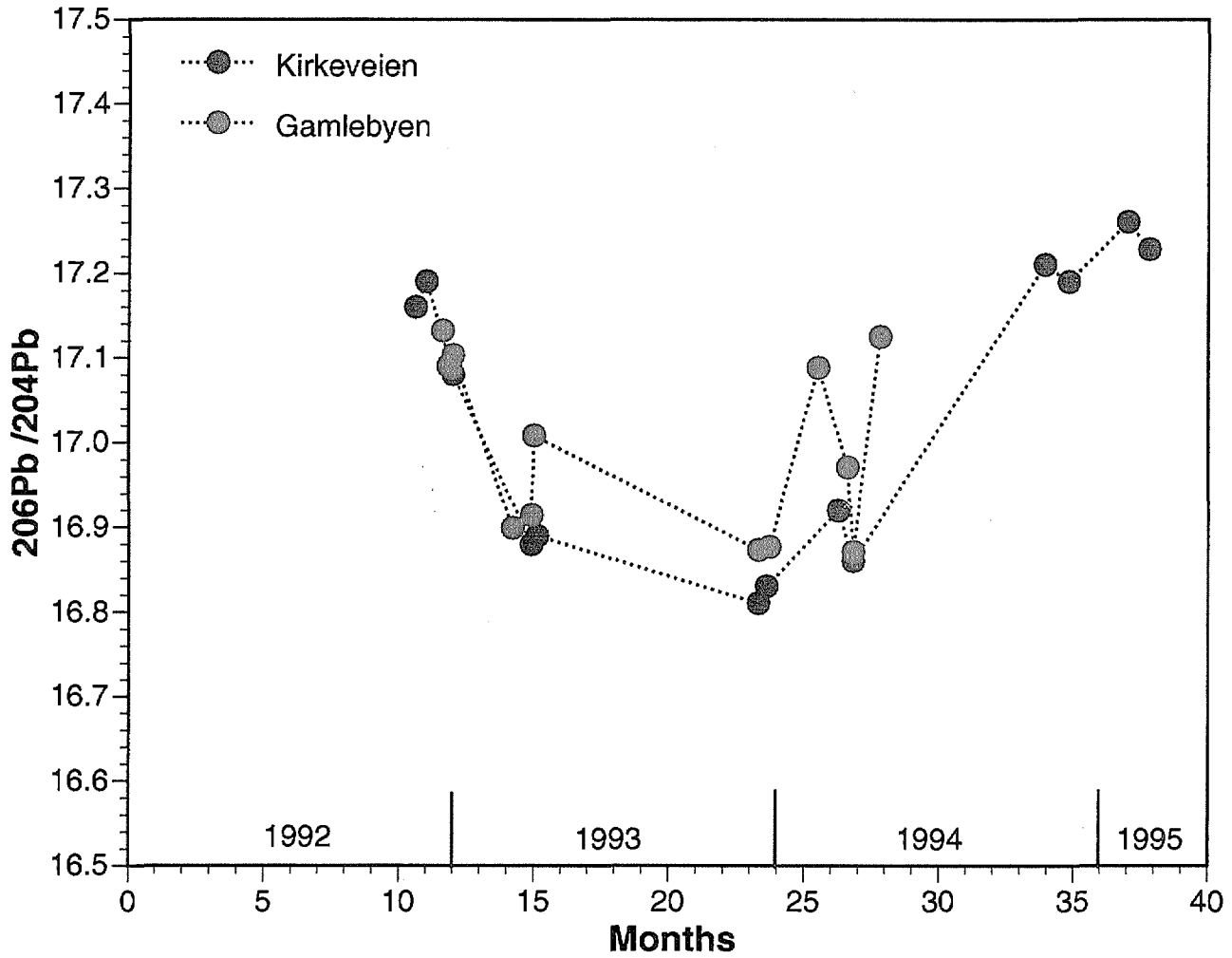


Figure 3. Pb isotope ratios for air filters collected at two sites in Oslo town between 1992 and 1995.

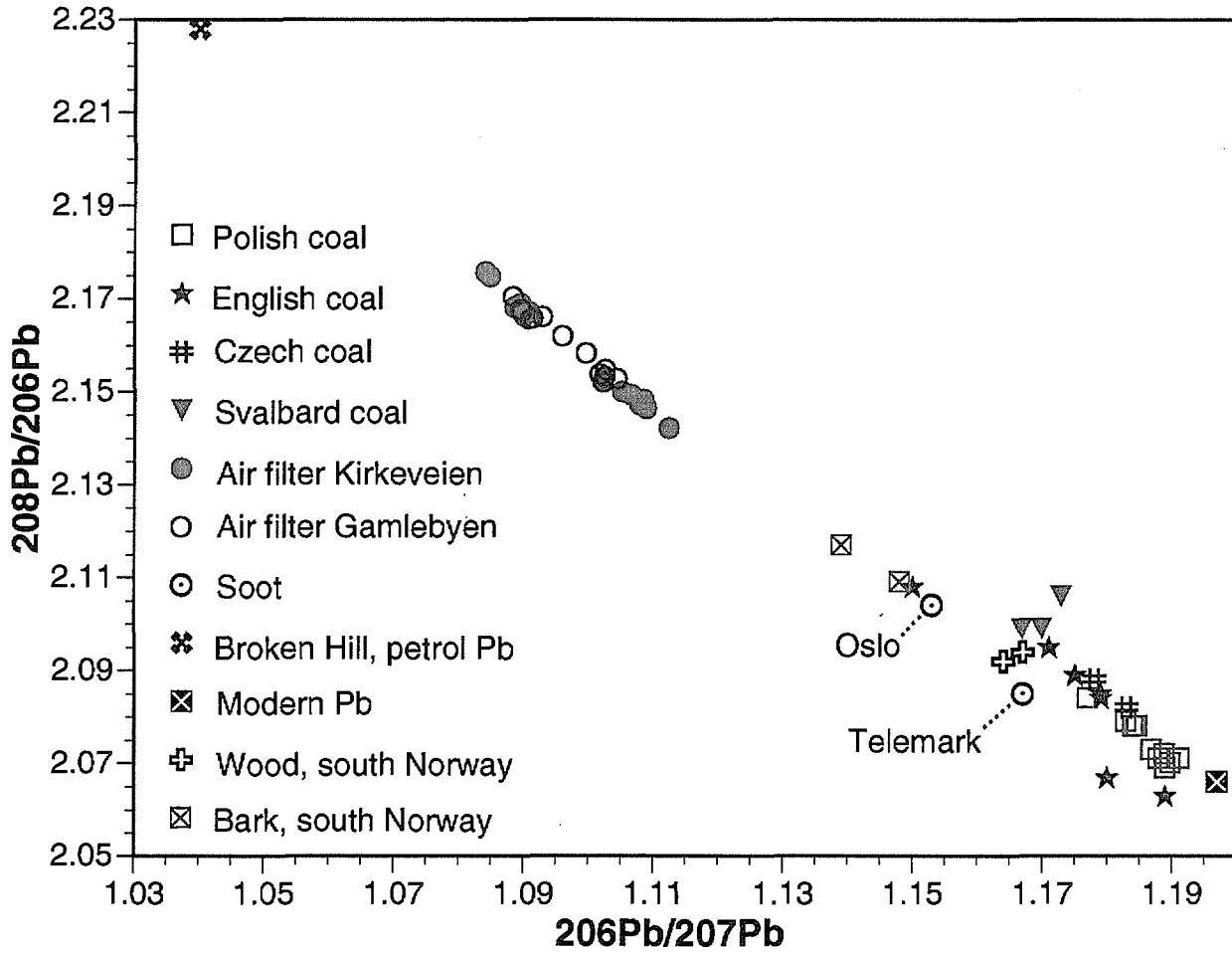


Figure 4. Pb ratios for air filter and different sources for Pb.

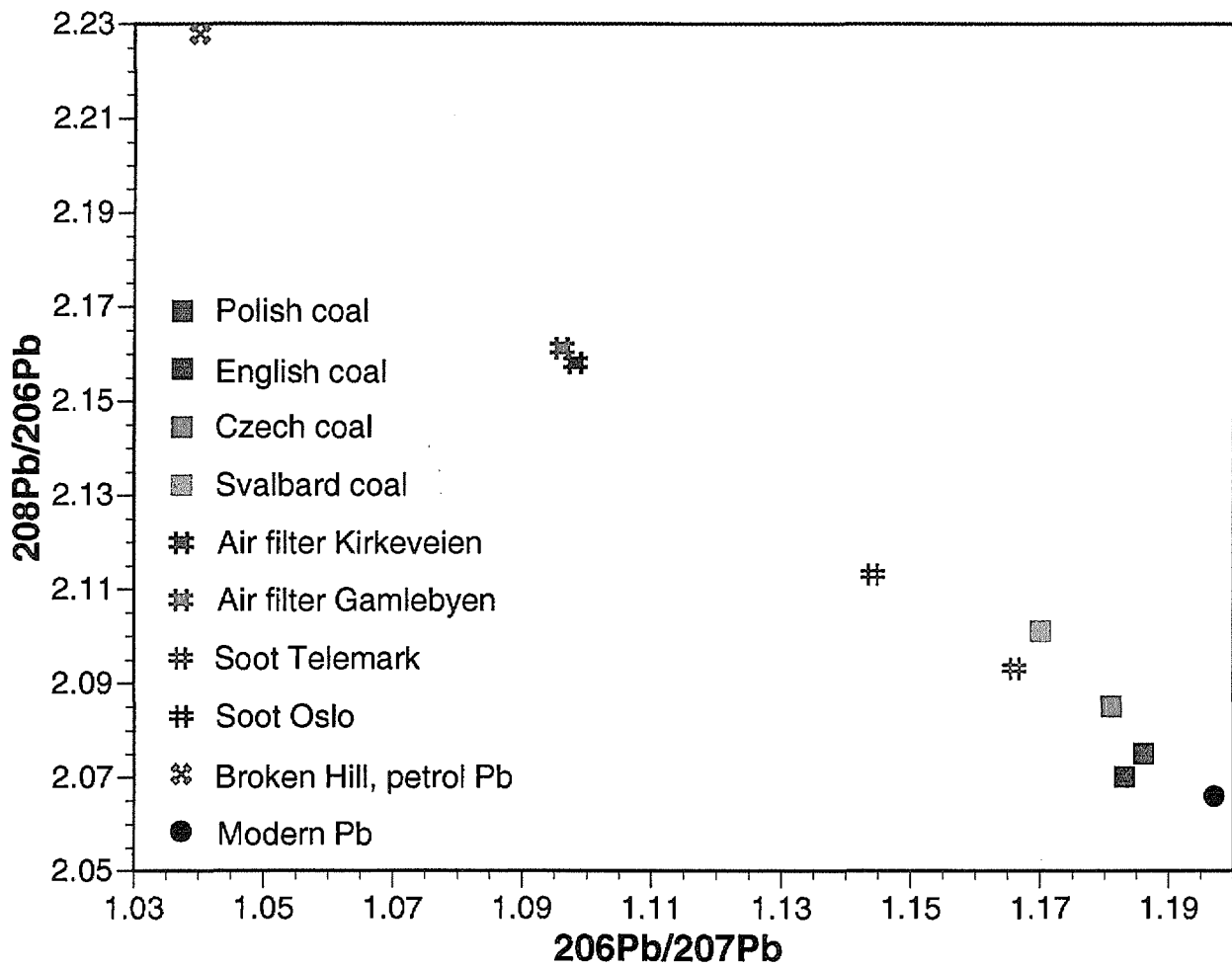


Figure 5. Mean of Pb ratios from Figure 4.

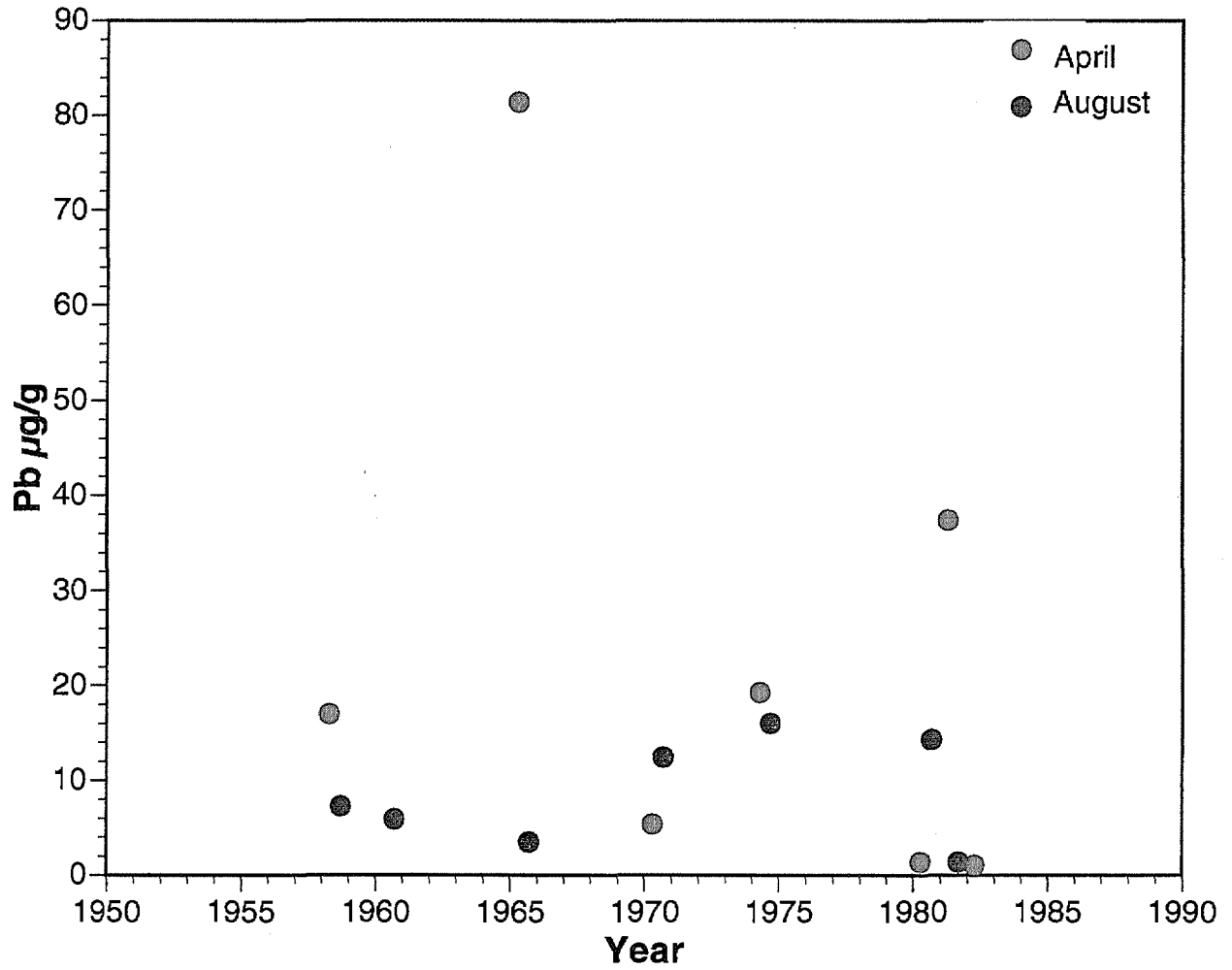


Figure 6. Pb concentration on air filter from Gardermoen sampled in April and August between 1958 and 1982

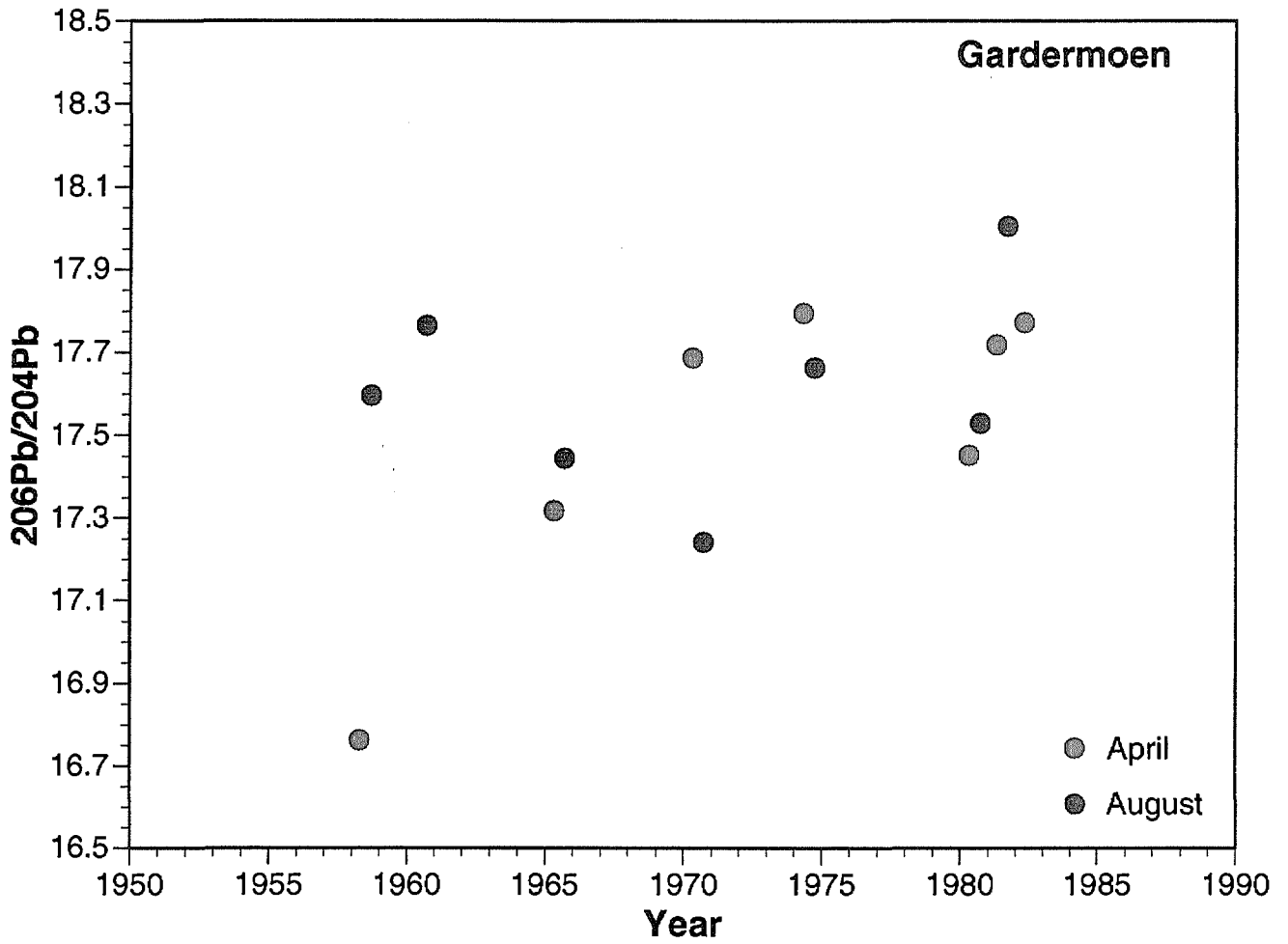


Figure 7. Pb ratio vs. time for air filter from Gardermoen sampled between 1958 and 1982.

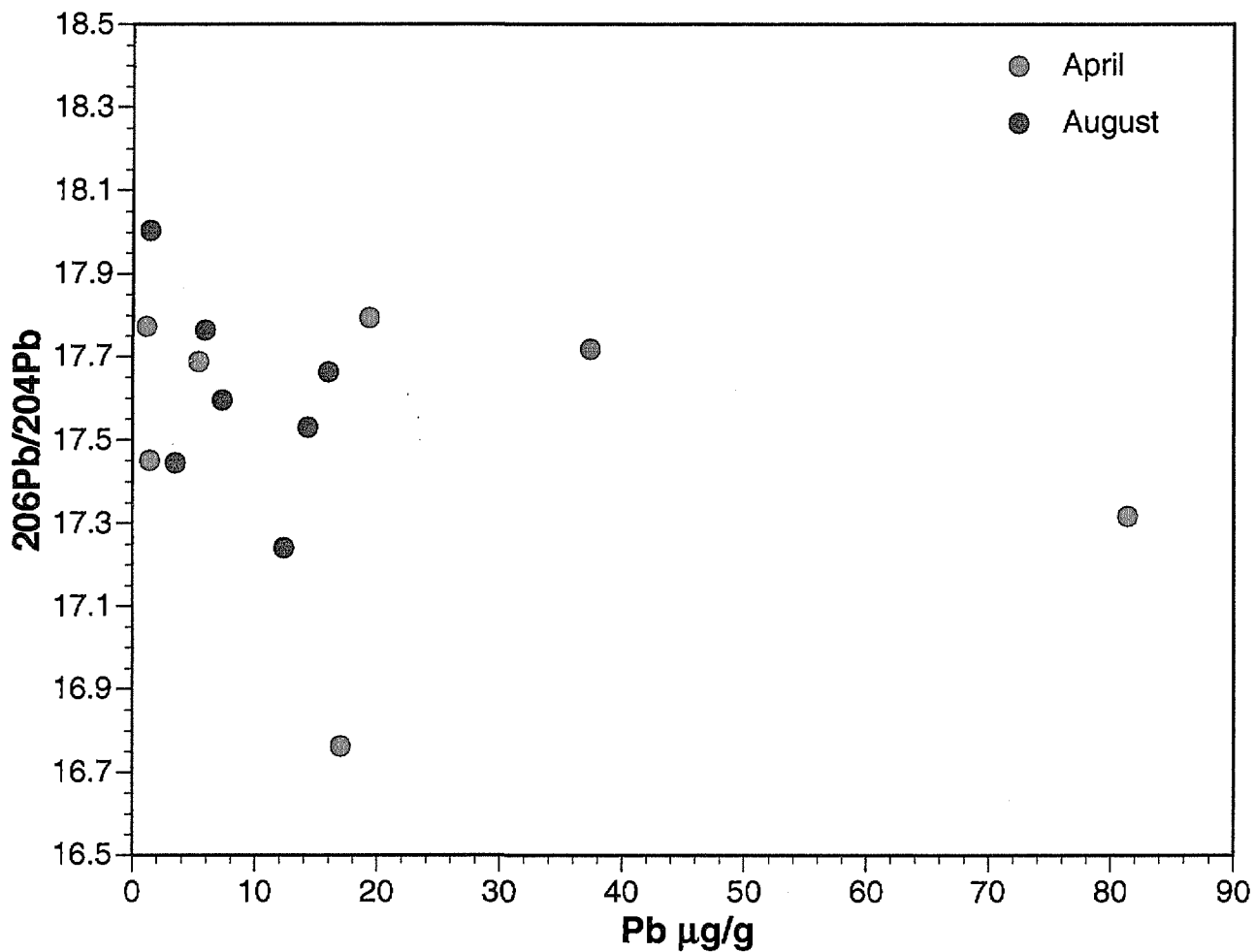


Figure 8. Pb ratio vs. Pb concentration for air filter from Gardermoen sampled between 1958 and 1982.