An assessment of global and regional emissions of trace metals to the atmosphere from anthropogenic sources worldwide

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Abstract: An accurate and complete emission inventory for atmospheric trace metals on a global scale is needed for both modeler community and policy makers to assess the current level of environmental contamination by these pollutants, major emission sources and source regions, and the contribution of the atmospheric pathway to the contamination of terrestrial and aquatic environments. Major progress has been made in assessing emissions of trace metals in various countries and even regions, e.g., Europe, since the first global emission estimate for these pollutants was made by Nriagu and Pacyna (1988). These improved national and regional emission inventories have been used in this work to assess the global trace metal emissions from anthropogenic sources in the mid-1990s. The results of this work conclude that stationary fossil fuel combustion continues to be the major source of Cr, Hg, Mn, Sb, Se, Sn, and Tl with respect to the coal combustion and the major source of Ni and V with respect to oil combustion. Combustion of leaded, low-leaded, and unleaded gasoline continues to be the major source of atmospheric Pb emissions. The third major source of trace metals is non-ferrous metal production, which is the largest source of atmospheric As, Cd, Cu, In, and Zn. The largest anthropogenic emissions of atmospheric trace metals were estimated in Asia. This can be explained by growing demands for energy in the region and increasing industrial production. As a result, the Asian emissions are not only larger than the emissions on other continents, but also show an increasing trend. Another factor contributing to high emissions in Asia is the efficiency of emission control, which is lower than in Europe and North America. Concerning the two latter continents, emissions of trace metals show a decreasing tendency over the last two decades.

Key words: anthropogenic sources, atmospheric emissions, trace metals, global emission inventory.

Résumé : Pour les constructeurs de modèles de communaut és aussi bien que pour ceux qui élaborent des politiques, il serait nécessaire de pouvoir disposer d'un inventaire complet et pr écis des métaux traces atmosphériques, afin de pouvoir évaluer, le degré actuel de contamination environnementale par ces polluants, les principales sources d'émission et les régions sources, ainsi que la contribution via l'atmosphère à la contamination de l'environnement terrestre et aquatique. Il y a eu un progrès majeur dans l'évaluation des émissions de métaux traces dans divers pays et même régions, p. ex. en Europe, depuis la première évaluation globale de ces polluants effectuée par Nriagu et Pacyna en 1988. Ces

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inventaires nationaux et régionaux améliorés ont été utilisés dans ce travail afin d'évaluer les émissions globales anthropogènes de métaux traces, au milieu des années 1990. On peut en conclure que la combustion stationnaire de carburants fossiles continue d'être la source principale des Cr, Hg, Mn, Sb, Se, Sn et Ti reliée à la combustion du charbon et la principale source des Ni et V reliée à la combustion du pétrole. La combustion de gazoline sans plomb, faible en plomb et avec plomb est toujours la principale source d'émissions atmosphériques de plomb. La troisième source majeure de métaux traces est liée à la production des métaux non-ferreux, qui constitue la source la plus importante des As, Cd, Cu, In et Zn. Les émissions anthropogènes de métaux traces les plus importantes se retrouvent en Asie. Ceci peut s'expliquer par des demandes croissantes d'énergie dans la région et une production industrielle accrue. Il en résulte que les émissions asiatiques ne sont pas seulement plus importantes que les émissions sur les autres continents, mais qu'elles tendent également à s'accroître. Un autre facteur contribue aux fortes émissions en Asie ; il s'agit de l'efficacit é plus faible de la maîtrise des émissions qu'en Europe et en Amérique du Nord. Pour ce qui est de ces deux derniers continents, les émissions de métaux traces montrent une tendance à décroître au cours des deux dernières décades.

Mots clés : sources anthropogènes, émissions atmosphériques, métaux traces, inventaire global des émissions.

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1. Introduction

Trace metals may create adverse effects on the environment and human health depending upon their bioavailability and toxicity in various environmental compartments. During the last three decades a number of studies have been carried out to assess the fate and behavior of these metals in the environment, as well as their environmental effects. The results of these studies have been reviewed during the preparation of the substantiation documents for the United Nation (UN) Economic Commission for Europe (ECE) protocol on emission reductions of trace metals (e.g., UN ECE 1994). These results can be summarized as follows.

Many trace metals are ubiquitous in various raw materials, such as fossil fuels and metal ores, as well as in industrial products. Some trace metals evaporate entirely or partially from raw materials during the high-temperature production of industrial goods, combustion of fuels, and incineration of municipal and industrial wastes, entering the ambient air with exhaust gases. Releases to other environmental compartments (e.g., spills to water bodies, landfills, sewage lagoons, and holding ponds) may also result in volatilization and entrainment of several trace metals. After being emitted to the atmosphere, trace metals are subject to transport within air masses and migration through the ecosystem, which cause perturbations of their geochemical cycles not only on a local scale but also on regional and even global scale.

Concentrations of many metals measured at remote locations worldwide are often higher than the concentrations expected from their natural occurrence in the terrestrial and aquatic environment (Nriagu and Davidson 1986). Emissions of trace metals during various human activities, mostly on fine particles, are assumed to be the major cause of this concentration increase. The increase in concentration of a given metal in an air sample, measured in certain reference material such as crustal rocks or soils, in relation to a certain reference metal such as Al, Ti, or Sc can be defined as an enrichment factor (EF) of this metal. Most metals can be locally enriched. Some of them also show enrichment on regional and global scales. Episodic long-range transport of pollutants within air masses result in the enrichment of metal concentrations far from source regions, e.g., in the Arctic.

Deposition of trace metals in areas surrounding emission sources, as well as "en route" deposition during transport, has reached levels that in certain regions have exceeded the maximum permissible values on soils and fresh water ecosystems. After entering the terrestrial and aquatic environments in the emission regions, as well as far away from them, trace metals can accumulate in soils. Uptake of trace metals by terrestrial and aquatic organisms and their metabolism may also lead to bioaccumulation of certain trace metals in the environment, including Cd, Se, and Hg (AMAP 1999).

In general, the metal threat to the environment has been evaluated in terms of ecology, health, economics, and quality of life. It is beyond the scope of this paper to analyze these evaluation factors in detail. However, a very brief summary can be offered.

Effects of trace metal exposure have been recorded in forest waters in the vicinity of strong point sources, as well as at remote locations, due to long-range transport of these pollutants within air masses. The threats to the terrestrial environment, lake and sediments, fresh water and marine ecosystem, marine birds and mammals, and human health in the Arctic have been assessed during the first phase of the Arctic Monitoring and Assessment Programme (AMAP 1997). Major concern focused upon Hg and Cd, while Pb was considered to pose less threat. The Hg and Cd levels were considered high enough to raise public health concerns in the region (AMAP 1997). Studies carried out in the vicinity of large sources of trace metal emissions, e.g., in the Upper Silesia in Poland, concluded that high levels of Cd and Pb in the environment cause an increase of spontaneous abortion in women during the first 3 months of pregnancy (Krasnodebski 1998).[2]

Several studies pointed out the synergistic effect of various metals and acid deposition compounds (UN ECE 1994). Soil acidification may significantly enhance concentrations of various metals, including Cd and Zn, in soil solution leading to the increased availability of these pollutants to terrestrial and aquatic ecosystems (Pacyna 1998; AMAP 1999).

2. Objectives

Accurate and complete information on sources and releases is of vital importance for modeling the atmospheric and riverine transport of contaminants at all distances (on local, regional, and global scale)

[2] Krasnodebski, J. 1998. The impact of heavy metal contamination of the environment on the health of mothers and fetuses during the first threemester of pregnancy, personal communication on unpublished data from Upper Silesia in Poland.

and for assessing the impact of these contaminants on the environment and human health. The overall goal of this paper is to revise the information on global emissions of trace metals from anthropogenic sources, presented for the 1983 reference year by Nriagu and Pacyna (1988), and to propose a global emission inventory for the present time (the mid-1990s).

The need for such a paper is substantiated by the following:

- accurate and complete as possible global and regional emission inventories for atmospheric trace metals are urgently needed in view of the implementation of the recent international emission reduction agreements for trace metals, particularly the UN ECE protocol on reduction of trace metal emissions and the AMAP agreement on reduction of the contamination of the Arctic environment
- since the first assessment of anthropogenic emissions by Nriagu and Pacyna (1988), major improvement of emission control has recently been made in various industries and the consumption of low-leaded and unleaded gasoline in many countries has increased
- an increase in demand for electricity and heat based on combustion of fossil fuels, particularly coal, has been also observed in many countries during the last 15 years, particularly in Asia
- several measurements have been carried out during the last few years to assess the re-emission rate of trace metals, particularly Hg, from terrestrial and aquatic ecosystem surfaces resulting in better and more accurate input data for the assessment of emissions of these pollutants

The 1983 emission data and presentation of the emission data for the 1990s have been revised using (1) the information on emissions received from national experts in a number of the European countries and the United States, and (2) the estimates of emissions for the rest of the countries, carried out within the reported project by the authors of the report. Such method of work required a number of assumptions to be made and resulted in a degree of uncertainty of the presented emission estimates. These assumptions and uncertainty are discussed later in this paper.

Ideally, the assessment of trace metal emissions should be made on the basis of emission measurements, at least for major point sources of emissions, such as large power plants, smelters, and waste incinerators. Unfortunately, these measurements are carried out sporadically and are limited mostly to the measurements in non-ferrous metal smelters and large electric power plants. The results of these measurements cannot be easily extended to cover the whole source category or transferred from one country to another. The vast majority of emissions data summarized and presented in this paper are estimates generated either by national experts or the authors of this report.

3. Two decades of emission inventorying for trace metals

During the last two decades there has been great progress in defining major anthropogenic sources of Cd, Pb, Hg, and other trace metals in Europe and in other parts of the world (Pacyna et al. 1995; Olendrzynski et al. 1996; Pirrrone et al. 1996; Pacyna and Pacyna 1999). It was concluded that high-temperature processes, such as coal and oil combustion in electric power stations and heat and industrial plants, gasoline combustion, roasting and smelting of ores in non-ferrous metal smelters, melting operations in ferrous foundries, refuse incineration, and kiln operations in cement plants emit various trace metals, which enter the atmosphere and the aquatic and terrestrial ecosystems. Practically every industry discharges one trace metal or another into these ecosystems.

The first quantitative worldwide estimate of the annual industrial input of 16 trace metals into the air, soil, and water at the beginning of the 1980s was published by Nriagu and Pacyna (1988). Pyrometallurgical processes in the primary non-ferrous metal industries were found to be the major source of atmospheric As, Cd, Cu, In, Sb, and Zn, and an important source of Pb and Se. Combustion of coal in electric power plants and industrial, commercial, and residential burners was the major source of anthropogenic Hg, Mo, and Se and a significant source of As, Cr, Mn, Sb, and Tl. Combustion of

oil for the same purpose was the most important source of V and Ni. Combustion of leaded gasoline was estimated to be the major source of Pb. Atmospheric Cr and Mn were derived primarily from the iron and steel industry. The only updates of the above-mentioned global emission inventory have been prepared for Pb (Pacyna et al. 1995) and Hg (Pacyna and Pacyna 1996), with emission data calculated for the conditions at the beginning of the 1990s.

There were several attempts to assess atmospheric emissions of trace metals from anthropogenic sources on a regional (continental) scale during the past two decades. One of the first attempts to estimate atmospheric emissions of trace metals from anthropogenic sources in Europe was completed at the beginning of the 1980s (Pacyna 1984). This work presented information on emissions of as many as 16 trace metals, similar to those reviewed on a global scale by Nriagu and Pacyna (1988). European inventories prepared before the work by Pacyna (1984), mostly within the European Commission programs, have dealt with either a single metal (e.g., Cd or Pb) or a given source category (e.g., combustion of fossil fuels).

The above-mentioned European-wide emission survey was later updated, completed, gridded (Axenfeld et al. 1992), and applied in long-range transport models to study deposition of trace metals in Europe and at remote locations, such as the Arctic (a review by Pacyna (1994)).

In 1989 the UN ECE established a Task Force on Heavy Metals. One of the major objectives of the Task Force was to collect information on emissions. National response to the request for creating a database for emissions of trace metals in the UN ECE region was unsatisfactory at the beginning of the 1990s. At the same time, an update of emission data for Pb, Cu, and Cd for 1989–1990 has been made in connection with the Dutch project on a European Soil and Sea Quality and Atmospheric Deposition of Selected Substances (ESQUAD) (Berdowski et al. 1994).

Recently, several European countries have prepared national emission inventories for trace metals emitted from anthropogenic sources and have started to report them to the UN ECE European Monitoring and Evaluation Programme (EMEP) (UN ECE 1999*a*) in connection with the UN ECE Protocol on Heavy Metals, signed in June 1998. The objective of this protocol is to control emissions of trace metals caused by anthropogenic activities that are subject to long-range transboundary atmospheric transport and are likely to have significant adverse effects on human health or the environment (UN ECE 1998).

The Oslo and Paris Commissions (OSPARCom) program, concerning the transport of pollutants to the North Sea, has also collected information on atmospheric emissions of trace metals from anthropogenic sources in Europe since the beginning of the 1980s. In 1992 the Convention for the Marine Environment of the North-east Atlantic replaced the Paris Commission formed in 1974. In 1997 an inventory of the European emissions was prepared within the Convention for the reference year 1990 on the basis of the work within the ESQUAD project (Berdowski et al. 1997).

A set of estimates of current, past, and future emissions of As, Cd, Pb, and Zn in Europe has been prepared for the International Institute for Applied Systems Analysis (IIASA) with current data regarded as those at the beginning of the 1990s (Pacyna 1996). This work has been updated for Pb emissions in 1995 for the German GKSS Research Centre (Pacyna and Pacyna 1999).

First estimates of atmospheric emissions of As, Cd, Cu, Cr, Mn, Ni, Pb, Sb, Se, V, and Zn from anthropogenic sources in 12 major source regions in the former Soviet Union were prepared by Pacyna on the basis of emission factors and statistical data for the reference year 1979–1980 (NILU 1984). Emissions from non-ferrous and ferrous metal production, fossil fuel combustion, and gasoline combustion were estimated to dominate the total emissions in these regions. Only recently has this old emission inventory been updated and improved by studies carried out by Tsibulski et al. (1996), Gromov (1996) and Boyd et al. (1998).

Emission data for trace metals (except for Pb) from anthropogenic sources in the United States are much less comprehensive than those for the criteria air pollutants, such as sulfur dioxide, nitrogen oxides, carbon monoxide, volatile organic compounds (excluding certain nonreactive organic compounds), particulate matter less than 10 μm, lead, and total suspended particulate matter. There are two different

programs in the United States collecting information on emissions of toxics: Toxic Release Inventory (TRI) estimates (currently for over 300 chemicals in 20 chemical categories) submitted annually since 1987 to EPA by certain manufacturing facilities and national inventories for specific pollutants prepared by EPA to support special studies called for by the 1990 Clean Air Act Amendment (CAAA).

The EPA inventories are more comprehensive than the TRI in that they attempt to identify and quantify all source categories and air emissions of toxics, whether from manufacturing facilities, commercial facilities, mobile sources, or residential and consumer sources. These inventories also include emissions from facilities with fewer than 10 employees and emissions from sources with very low concentrations of toxics, which are exempted from the TRI reporting requirements.

Mercury and cadmium are the two trace metals for which national emissions have been prepared in the United States, in addition to Pb, which is a criteria pollutant. Mercury is one of the seven pollutants specified in the section of the Clean Air Act (CAA) that requires the EPA to identify source categories and sub-categories of Hazardous Air Pollutants (HAPs) in urban areas that pose a threat to human health. Emission estimates for both Hg and Cd were prepared within the Locating and Estimating (L&E) document series (US EPA 1993*a*, 1993*b* for Hg and Cd, respectively). The L&E documents provide a compilation of available information on the sources and emissions of specific toxic air pollutants, including the information on emission factors. There are also more recent studies on specific trace metals, particularly Hg (Porcella et al. 1996).

Canadian emission data for trace metals have been published by Environment Canada starting with data for 1982 (Jacques 1987). Almost no data exist on emissions of trace metals from anthropogenic sources in Mexico. Porcella et al. (1996) have extrapolated values from the U.S. emission survey to assess the Mexican emissions of Hg.

A few emission inventory programs have been carried out in Asia. Lead has been the most studied metal. Major revision of results from these programs has been made during the International Symposium on Emission Inventory and Prevention Technology for Atmospheric Environment, held in Japan (ITIT 1992). The emission estimates for Japan, Thailand, China, and India in the mid-1980s were presented. It is not clear whether any updates and modifications of these emission inventories have been made recently.

Emission inventories for lead have been also carried out in some African and South American countries (a review by Pacyna et al. 1995; Nriagu et al. 1996). Releases of Hg during gold and silver mining were reviewed by Lacerda (1997).

Information presented in the above-mentioned inventories has been used in this work to update the 1983 global emission estimates and to estimate and discuss global emissions of trace metals in the mid-1990s presented in section 4 of the report.

4. Assessment of global regional emission estimates for the year 1995

Two approaches were undertaken for presentation of global anthropogenic emissions of trace metals in the reference year 1995: (1) collection of emission data from countries where such data were estimated by national emission experts and (2) estimates of emissions on the basis of emission factors and statistical data on the production of industrial goods and (or) the consumption of raw materials. These estimates were carried out by the authors of this report for the countries with no national estimates.

4.1. Assessment of anthropogenic trace metal emission data supplied by national experts

It is often perceived in international emission inventories that data estimated or measured by local–national emission experts are more complete and accurate for a given region than the estimates by international emission experts. Local emission experts would be aware in more degree of details about raw materials used in their regions, the application of certain type of industrial–combustion technologies, and emission control equipment. Therefore, it is very important for experts assessing global emissions to obtain as much available information from local–national experts.

The following data were received from national emissions experts from 20 countries. Details on these data are available from reports presented in Appendix A.

The emission data received from national authorities have then been checked for completeness and comparability. The completeness of data regarded mainly the inclusion of all major source categories that may emit trace metals to the atmosphere. No major omissions have been detected. All major source categories in all countries reporting the emission data were included in this reporting.

It is very difficult to verify the data obtained from national authorities in various countries in Europe. The following approach has been taken. Information on emissions of trace metals from various sources was brought together with statistics on the production of industrial goods and (or) the consumption of raw materials, and these two sets of data were used to calculate emission factors. Emission factors calculated in this way were then compared with emission factors reported in the Joint EMEP/ CORINAIR Atmospheric Emission Inventory Guidebook (UN ECE 1999*b*). For the majority of cases, emission factors estimated on the basis of national emission data reported to the project were within the range of emission factors proposed in the Guidebook. This check was proof that the estimates from various countries are comparable and thus can be accepted for the reported project.

All data received from above-mentioned 20 countries were accepted in this work as representative for national emission inventories for these countries.

4.2. Emission estimates for countries with no national data available

Emission estimates have been performed in this work for the countries where no national emission data were available. These estimates have been based on the statistical information on the consumption of raw material and the production of industrial goods, available from international and national statistical yearbooks and the emission factor data. The emission factors used in the estimates are discussed below together with the emission estimate results for each source category, separately.

4.3. Assessment of the 1995 global emissions from individual source categories

4.3.1. Emissions from the production of electricity and heat in 1995

The emission estimates for various trace metals emitted during the production of electricity and heat in 1995 are presented in Table 1. These emission data consist of information received from various national authorities in Europe (as mentioned above) and the United States and Canada with regard to emissions of Hg and Pb. For the remaining countries, emission estimates were carried out on the basis of information on the energy production from the Statistical Yearbook (UN 1997) and emission factors. Average values of the 1995 emission factors used to estimate trace metal emissions from stationary fossil fuel combustion in countries with no national emission estimates available for the authors are presented in Table 2. These factors are in the lowest part of the range of emission factors suggested by Nriagu and Pacyna (1988) for the 1983 emission estimates. The 1995 emission factors used in the reported work are within the range of emission factors suggested by the UN Economic Commission for Europe (ECE) Emission Inventory Task Force for use in the emission estimates within the UN ECE countries. The UN ECE emission factors are available in the Joint EMEP/ CORINAIR Atmospheric Emission Inventory Guidebook (UN ECE 1999*b*). The emission factors in Table 2 are proposed in the Guidebook for use in the cases of power plants with standard emission control equipment.

The largest emissions of trace metals from stationary fossil fuel combustion have been estimated for Asia, accounting for 40 to 60% of the total emissions depending on the metal of concern. The only exception is the emission of Cd. The data provided by the US EPA indicate that the combustion of fuels in the United States alone contributed as much as 241.6 t to the total global emissions in 1990 (US EPA 1993*b*). It was assumed that similar emissions can be assigned as valid for the mid-1990s for the United States because of the similar amounts of coal and oil combustion in various years at the beginning of the 1990s. No major improvement of emission control technology in the US power plants was also assumed

Table 1. Emission of trace metals from combustion of fuels in stationary sources in 1995 (in tonnes) and comparison with the 1983 emission estimates by Nriagu and Pacyna (1988).

Continent	Hg	As	Cd	Cr	Cu	Mn	Mo	Ni	Pb	Sb	Se	Sn	Tl	Zn	V
Europe	186	143	109	1 999	1342	1 875	531	20 072	2 683	124	780	815	311	1 875	57 107
Africa	197	41	40	746	459	715	207	9 902	1 116	31	269	399	78	715	28 924
Asia	860	342	237	4 282	2989	3 974	1115	36 320	4 845	308	1731	1484	771	3 974	101 254
North America	105	234	263	2 342	1792	2 117	579	10 752	1 978	226	1030	453	564	2 117	26 615
South America	27	9	22	429	216	428	128	8 479	850	1	129	339	1	428	25 422
Australia and Oceania	100	40	20	347	283	308	82	585	218	40	162	27	99	308	762
1995 world total	1475	809	691	10 145	7081	9 417	2642	86 110	11 690	730	4101	3517	1824	9 417	240 084
1983 world total	2081	2037	673	12 683	7144	12 106	3268	40 833	10 577	1304	2312	4267	1130	13 259	84 045

Table 2. Average emission factors for trace metals used to estimate emissions from the stationary fossil fuel combustion, the cement production, and the pig iron and steel production.

	Fossil fuel combustion			
Trace metal	Coal (g/t coal)	Oil (g/t oil)	Cement production (g/t cement)	Pig iron and steel production (g/t steel)
As	0.2	0.02	0.2	0.5
Cd	0.1	0.05	0.01	0.1
Cr	1.7	1.0	1.0	4.0
Cu	1.4	0.5	—	0.2
Hg	0.5	0.06	0.1	0.04
Mn	1.5	1.0	—	1.5
Mo	0.4	0.3	—	—
Ni	2.0	20.0	0.1	0.05
Pb	1.0	2.0	0.2	1.5
Sb	0.2	—	—	0.01
Se	0.8	0.3	0.002	0.01
Sn	0.1	0.8	—	—
Tl	0.5	—	—	—
V	1.0	60.0	—	0.1
Zn	1.5	1.0	2.0	3.0

Table 3. The 10 largest emitter countries for Hg from stationary fossil fuel combustion in 1995 (emissions in tonnes).

No.	Country	Emission
1	China	495
2	India	117
3	Australia	97
4	Zaire	90
5	United States	77
6	South Africa	76
7	Russia	54
8	Japan	45
9	Korea (Rep. Dem.)	44
10	Kazakhstan	36

between 1990 and 1995. It should be noted that the emission of Cd in the United States reported by the US EPA is unusually high, almost 2.5 times higher than in China, the second largest emitter country for this metal from fossil fuel combustion in stationary sources.

Mercury is often considered as the most interesting trace metal emitted from combustion of fossil fuels in power plants and heat-producing units, particularly those burning coal. This is because a major part of Hg is emitted in a gaseous form, passing the control equipment and entering the atmosphere. Almost 1500 t of this element were emitted from fossil fuel combustion in stationary sources in 1995. The 10 largest emitter countries are presented in Table 3. China alone emits one third of the total global emissions of Hg from this source, followed by India. The Asian countries emit almost 60% of the total emissions.

Vanadium and nickel are the two trace metals emitted in large amounts during the combustion of oil. The emission ratio between these two metals resembles the ratio of the concentrations in crude oil,

as well as in residual oil used in power plants and heat-producing units.

An interesting comparison can be made between the 1995 emission data and the emission estimates for the reference year 1983 (Table 1). For trace metals emitted mostly during coal combustion, namely Hg, As, Cd, Cr, Cu, Mn, Mo, Pb, Sb, Sn, and Zn, emissions in 1995 are either similar or lower than in 1983. Detailed information on country by country emissions in 1983 is not available for all countries, therefore it is difficult to make direct comparison. The pattern of coal combustion indicates that while the consumption of coal in Europe and North America did not change substantially, the demand for coal-based energy in the Asian countries rose quite significantly between the beginning of the 1980s and the mid-1990s. Major improvement of the efficiency of emission control for fine particles has been observed in Europe during the 1980s resulting in the decrease of emissions of trace metals in the period of comparison. Another factor contributing to the decrease of trace metal emissions in Europe was a change from coal combustion to oil and natural gas combustion, as well as from fossil fuel combustion to nuclear energy and to larger use of hydro power in some countries. The detailed analysis of these changes is, however, beyond the scope of this paper.

The aforementioned pattern of trace metal emission decrease in Europe and North America has been compensated by the growing electricity demand in Asia. As a result, the emissions of Cd, Cr, Cu, Mn, Mo, Pb, and Sn from stationary fossil fuel combustion have not change significantly on a global basis between the beginning of the 1980s and the mid-1990s.

Estimates of lower emissions of Hg, As, and Zn in 1995 and higher emissions of Se and Tl are probably due to more accurate and complete information on emission factors for these compounds used to estimate the 1995 emissions. They do not follow the pattern of coal use and the emission control improvements.

An interesting comparison has been noted for the emissions of V and Ni, thus metals emitted mainly during oil combustion. The 1995 emissions are 2 to 3 times higher than the 1983 emissions, nicely resembling the factor of 2.2 increase in oil combustion to meet electricity and heat demands between the beginning of the 1980s and the mid-1990s. Indeed, the consumption of oil and the metal content of oil are the main factors affecting the amount of V and Ni emissions to the atmosphere from oil combustion in power plants. Most of the oil-firing power plants are not equipped with any dedusting installations. It can also be assumed that the content of Ni and V in crude oil has not changed significantly in the period of comparison. Thus, the main explanation of the more than double increase of V and Ni emissions from oil-firing power plants would be increased production of electricity and heat worldwide.

4.3.2. Lead emissions from vehicular traffic

Combustion of all kinds of gasoline results in emission of Pb to the atmosphere. There are also other trace metals that can be emitted from this source but these emissions are negligible. Obviously, the largest Pb emissions are from leaded gasoline combustion.

Accurate information on the amount of leaded, low-leaded, and unleaded gasoline used in a given country is of primary importance for the estimates of atmospheric emissions of the metal. This information, as well as the information on national emissions of Pb from vehicular traffic in 1995, was available from national authorities in the European countries, the United States, and Canada. For other countries, emission estimates have been carried out by the authors. It should be noted that no information was obtained on the consumption of leaded, low-leaded, and unleaded gasoline in these countries, nor on the lead content of gasoline in 1995. In such circumstances an assumption has been made that for the countries other than the European countries, the United States, and Canada, the Pb emission factors for 1995 will be at a level of those estimated by Pacyna et al. (1995) for 1989. The results of these estimates are presented in Table 4.

The largest contribution, 43–44% of the worldwide vehicular emissions of Pb, comes from the combustion of gasoline in Asia. The largest contributions from individual countries come from European Russia, China, and Mexico, each emitting over 8500 t of Pb in 1995.

Table 4. Worldwide emissions of Pb from mobile sources in 1995 (in tonnes).

Continent	Minimum	Maximum
Europe	19 507	19 507
Africa	6 852	11 992
Asia	32 996	44 293
North America	10 414	15 780
South America	4 866	7 270
Australia	2 000	2 000
World total	76 635	100 842

The average worldwide Pb emission in 1995 was estimated at 88 700 t and can be compared with the estimates of average emissions of 116 700 t in 1989 (Pacyna et al. 1995) and with the estimates of average emissions of 248 000 t in 1983 (Nriagu and Pacyna 1988). This emission decrease in the period from the beginning of the 1980s to the mid-1990s corresponds well with the implementation of environmental strategies of Pb emission reductions through the introduction of low-leaded and unleaded gasoline in Europe, the United States, and Canada.

4.3.3. Emissions from non-ferrous metal production

Emissions from non-ferrous metal industry contributed most to 1983 worldwide emissions of As, Cd, Cr, Cu, In, Mn, and Zn (Nriagu and Pacyna 1988). The estimates of emissions from this source category have also caused the largest controversy in the literature concerning the value of emission factors and then the amounts of emissions in various smelters, technologies, countries, etc. (Skeaff and Dubreuil 1997). Indeed, the best solution for assessing the emission of trace metals from smelters is to measure these emissions, particularly the stack emissions. Such measurements have been carried out in the European, Canadian, U.S., and Australian smelters, but they are largely lacking in other countries. In the latter case emission factors need to be used for the emission estimates.

Since the publication of the first global emission inventory for trace metals by Nriagu and Pacyna (1988) a number of improvements have been made in the non-ferrous metal industry, including the introduction of very efficient control equipment to control both particulate and gaseous pollutants. To review the degree of these improvements, it was necessary to contact the non-ferrous metal industry. The following smelters submitted information to the authors: Cominco Ltd in Canada, Hudson Bay Mining and Smelting Co. Limited in Canada, Kennecott Utah Copper Corporation in the United States, Huttenwerke Kayser AG in Germany, Berzelius Metall GmbH in Germany, Norddeutsche Affinerie in Germany, and Metaleurop Weser Blei GmbH in Germany.

The information received from the above smelters, national environment protection authorities in the European countries, Canada, and the United States, as well as the estimates by the authors for other countries was brought together and presented in a form of trace metal emissions to the atmosphere from non-ferrous metal industry in the mid-1990s. The results are presented in Tables 5 through 7 for primary and secondary copper and nickel production, primary and secondary lead production, and primary and secondary zinc production, respectively. The copper – nickel industry emits larger quantities of trace metals (except for Hg and Zn), compared to the lead and zinc industry. Obviously, primary production processes emit most of the emissions from non-ferrous metal manufacturing.

Summary of emissions of trace metals from the non-ferrous metal industry is presented in Table 8. The largest emissions were estimated for the non-ferrous metal industry in Asia, followed by the smelters in South America. The emissions for these two continents were estimated in the reported project on the basis of emission factors and statistical data on the metal production. The average emission factors used

Table 5. Emissions of trace metals from the copper production in 1995: (*a*) primary production, and (*b*) secondary production (in tonnes).

Continent	As	Cd	Cu	Hg	In	Mn	Ni	Pb	Sb	Se	Sn	Zn	V
(*a*) *Primary production*													
Europe	185	92	555		2	2	277		19	19	19	370	9
Africa	260	104	1 562	5	2	5	781	521	26	26	26	260	5
Asia	1 593	637	9 555	32	13	32	4778	3 185	159	159	159	1593	32
North America	252	126	754	14	3	3	377	754	25	25	25	503	13
South America	868	347	5 207	17	7	17	2604	1 736	87	87	87	868	17
Australia and Oceania	25	13	75	1			37	75	3	3	3	50	1
World total	3 183	1319	17 708	69	27	59	8854	6 271	319	319	319	3644	77
1983 emission (average)	10 625	2550	22 525	122	21	2550	7650	16 575	1063	854	1063	6375	64
(*b*) *Secondary production*													
Europe	2	3	91				1		3			181	
Africa			1					1				1	
Asia			9					8				18	
North America	1	2	55				1	49	2			109	
South America			2					1				3	
Australia and Oceania			2					2				5	
World total	3	5	160				2	62	5			317	

Table 6. Emissions of trace metals from lead production in 1995: (*a*) primary production, and (*b*) secondary production (in tonnes).

Continent	As	Cd	Cu	Hg	Ni	Pb	Sb	Se	Zn
(*a*) *Primary production*									
Europe	2	8	4	2	4	3 341	15	8	15
Africa	1	1	1		1	195	7	3	8
Asia	12	17	12	3	12	2 298	86	40	92
North America	2	7	4	2	4	137	14	7	14
South America	1	2	1		1	203	8	4	8
Australia and Oceania	1	2	1	1	1	41	4	2	4
World total	19	37	23	8	23	6 215	134	64	141
1983 emission (average)	1170	117	273	12	166	21 450	293	293	332
(*b*) *Secondary production*									
Europe		2	1						17
Africa						5			1
Asia		1				20			4
North America	1	3	1			110			22
South America						5			1
Australia and Oceania						3			1
World total	1	6	2			143			46

Table 7. Emissions of trace metals from the zinc production in 1995: primary production and secondary production (in tonnes).

	Primary production									Secondary production
Continent	As	Cd	Cu	Hg	In	Pb	Sb	Se	Zn	Zn
Europe	57	108	13	14	7		29	22	3 237	123
Africa	6	25	6	3		61	2	2	1 228	
Asia	130	522	130	52	8	1304	39	39	26 082	3
North America	31	58	7	9	4	448	15	12	1 727	35
South America	19	77	19	8	1	191	6	6	3 827	2
Australia and Oceania	8	15	2	2	1	119	4	3	457	1
World total	251	805	177	88	21	2123	95	84	36 558	164
1983 emission (average)	460	2760	460		3	8510	69	161	64 400	

in these estimates are presented in Table 9. The emission factors for smelters in Asia, South America, and Africa were those considered in the UN ECE Joint EMEP/ CORINAIR Atmospheric Emission Inventory Guidebook (1999*b*) as representative for smelters with a standard emission control system. Statistical information on the production of non-ferrous metals was obtained from the World Bureau of Metal Statistics (WBMS 1999) and Industrial Commodity Statistics Yearbook 1996 (UN 1998).

Although most of the emission data for smelters in Europe, North America, and Australia were based on the information received directly from them or from their country national environmental protection authorities, in some cases emissions were calculated in this work. The average emission

Table 8. Emissions of trace metals from non-ferrous metals production in 1995: (*a*) primary production, and (*b*) secondary production (in tonnes).

Continent	As	Cd	Cu	Hg	In	Mn	Ni	Pb	Sb	Se	Sn	Zn	V	Cr
(*a*) *Primary production*														
Europe	245	208	572	15	8	2	281	3 341	62	48	19	3 622	9	
Africa	267	130	1 569	8	2	5	782	777	35	31	26	1 496	5	
Asia	1 735	1176	9 697	87	20	32	4789	6 787	285	239	159	27 767	32	
North America	284	191	765	25	6	3	381	1 340	54	44	25	2 244	13	
South America	888	425	5 228	25	8	17	2605	2 130	100	96	87	4 703	17	
Australia and Oceania	34	30	78	4	1		38	235	11	8	3	511	1	
World total	3 453	2160	17 909	164	45	59	8876	14 610	547	466	319	40 343	77	
1983 emission (average)	12 255	5427	23 258	134	24	2550	7816	46 535	1425	1308	1063	71 107	64	
(*b*) *Secondary production*														
Europe	2	5	92				1		3			321		
Africa			1					6				2		
Asia		1	9					28				25		
North America	2	5	56				1	159	2			167		
South America			2					7				7		
Australia and Oceania			2					5				7		
World total	4	11	162				2	205	5			529		
1983 emission (average)		11						765				855		11

factors applied for the calculation of smelter emissions in the European and North American countries are also presented in Table 9. More advanced control equipment was considered for these countries compared to the smelters in Asia, Africa, and South America. This consideration was given on the basis of comparisons between smelters described in a series of books on Non-Ferrous Metal Works of the World, published by Metal Bulletin Books.

The data in Table 8 also present a comparison with the 1983 emission estimates by Nriagu and Pacyna (1988). The 1995 emissions are lower by a factor of 2 to 3, except for Hg, In, Mn, Ni, and V. The decrease of emissions during the period from the beginning of the 1980s to the mid-1990s can be explained by a significant improvement of emission control efficiency for fine particles carrying trace metals in the European, Canadian, and U.S. smelters. Another explanation could be that 1983 data are entirely estimates, while more accurate information through the emission measurements was available for the 1995 data, as mentioned above.

The Hg emissions in 1995 are somewhat higher that the 1983 data, mostly because the 1983 data do not contain the information on Hg emissions from zinc production. Concerning the data for In, Mn, Ni, and V, the differences between the 1995 and 1983 estimates are probably because the 1983 Mn emission factors were overestimated, while the 1983 emission factors for In, Ni, and V were underestimated.

It should also be added that Hg is emitted during gold production. An earlier estimate of the Hg global emissions in 1990 by Pacyna and Pacyna (1996) indicates that as much as 325 t of Hg can be emitted from this process annually. More than half of these emissions occur in Africa. As no better estimates were available to the authors, the 1990 Hg emissions were accepted as valid also for the mid-1990s.

The emission factors used in the reported work fit very well within the range of emission factors proposed within the UN ECE Guidebook mentioned earlier in this report. The data from the Guidebook were tested and approved for use by a great number of emission experts in Europe and also in the United States. Therefore, the estimates presented in this paper are claimed to be more accurate than those used by Skeaff and Dubreuil (1997).

4.3.4. Emissions from iron and steel industry and cement production

Emissions of trace metals from the iron and steel manufacturing and cement production were estimated for the year 1994. As in the case of other source categories presented in this work, a number of European countries, the United States, and Canada provided their national emission values, which were used to prepare a global emission inventory reported here. Emissions for the remaining countries were estimated in this work. These estimates were performed on the basis of emission factors and statistical information on the pig iron and steel production and cement production. The average values of these factors are presented in Table 2. The production statistics were obtained from the UN Statistical Yearbook (UN 1997).

The emission estimates for the pig iron and steel production are reported in Table 10, and the cement industry in Table 11. The largest emissions were estimated for Asia and Europe. Particularly high emissions were estimated for Cr.

Comparison of the emission estimates in this work with the estimates for 1983 emissions indicates large differences. There are various reasons for these differences, which are very difficult to assess quantitatively. The first reason is the improvement of the emission control for fine particles during the period of comparison. Indeed, a major improvement of the ESP and multicyclone abatement efficiencies has occurred in Europe in the mid-1980s and in Asia at the beginning of the 1990s. The second reason is related to the quality of emission factors used in the 1983 estimates. The collection of information on emission factors by the authors for the UN ECE Joint EMEP/ CORINAIR Atmospheric Emission Inventory Guidebook (1999*b*) has proved that the 1983 emission factors could have been too high.

Mercury was not taken into account when preparing the 1983 emission inventory for the iron and steel industry and cement production. The same applies to Se from the cement production. Emissions

Table 9. Emission factors for non-ferrous metals by continent in 1995 (grams/tonne).

	Primary product Cu		Secondary product Cu	Primary product Pb		Secondary product Pb	Primary product Zn		Secondary product Zn
Element	Europe, N.A., Australia	Africa, Asia, S.A.	Europe, N.A., S.A., Australia, Africa, Asia	Europe, N.A., Australia	Africa, Asia, S.A.	Europe, N.A., S.A., Australia, Africa, Asia	Europe, N.A., Australia	Africa, Asia, S.A.	Europe, N.A., S.A., Australia, Africa, Asia
As	100.0	500.0	2.0	3.0	10.0	0.5	26.0	50.0	—
Cd	50.0	200.0	3.0	10.0	15.0	2.5	49.0	200.0	—
Cu	300.0	3000.0	100.0	5.0	10.0	1.0	6.0	50.0	—
Hg	5.6	10.0	—	3.0	3.0	—	7.6	20.0	—
In	1.0	4.0	—	—	—	—	3.0	3.0	—
Mn	1.0	10.0	—	—	—	—	—	—	—
Ni	150.0	1500.0	1.0	5.0	10.0	—	—	—	—
Pb	300.0	1000.0	90.0	200.0	2000.0	100.0	380.0	500.0	—
Sb	10.0	50.0	3.0	20.0	75.0	—	13.0	15.0	—
Se	10.0	50.0	—	10.0	35.0	—	10.0	15.0	—
Sn	10.0	50.0	—	—	—	—	—	—	—
Zn	200.0	500.0	200.0	20.0	80.0	20.0	1 466.0	10 000.0	270.0
V	5.0	10.0	—	—	—	—	—	—	—

Note: N.A., North America; S.A., South America.

Table 10. Emission of trace metals from pig iron and steel production in 1994 (in tonnes).

Continent	Hg	As	Cd	Cr	Cu	Mn	Ni	Pb	Sb	Se	Zn	V
Europe	10	130	26	1 037	52	389	13	2255	3	3	778	26
Africa	1	6	1	49	2	18	1	18			36	1
Asia	12	139	28	1 111	56	417	14	417	3	3	833	28
N. America	5	57	4	456	23	171	6	171	1	1	342	11
S. America	1	17	4	138	7	52	2	52			104	4
Australia and Oceania		4	1	34	2	13		13			25	1
World total	29	353	64	2 825	142	1 060	36	2926	7	7	2 118	71
1983 emission (average)		1418	156	15 620	1491	14 733	3568	7633	5	2	19 525	746

Table 11. Emissions of trace metals from cement production in 1994 (in tonnes).

Continent	Hg	As	Cd	Cr	Ni	Pb	Se	Tl	Zn
Europe	26	55	3	273	27	55	1		547
Africa	5	10	1	52	5	10			104
Asia	82	164	8	818	82	164	2		1635
North America	13	26	4	128	13	26			257
South America	6	11	1	56	6	11			111
Australia and Oceania	1	2		8	1	2			16
World total	133	268	17	1335	134	268	3		2670
1983 emission (average)		534	271	1335	490	7129		4005	9790

of Hg from cement production proved to be rather high, particularly from cement kilns in the Asian countries. In general, however, emissions of trace metals from the iron and steel industry and cement production are substantially lower than the emissions from non-ferrous metal industry and stationary fossil fuel combustion. Therefore, somewhat lower quality of emission factors expected for the iron and steel industry and cement production as compared to other major source categories will not have a significant impact on the quality of the estimates of total emissions of trace metals in the world.

4.3.5. Emissions from waste disposal

Emissions from the incineration of municipal wastes and sewage sludge are the most difficult to estimate because of a lack of information on the amounts of wastes incinerated in various countries and their composition. Thus, it is also difficult to assess the content of trace metals in the wastes.

The national emission estimates for the incineration of wastes have been available from only a few countries in Western Europe and the United States. These estimates were used directly in this work. Additionally, statistics on the amount of wastes incinerated were available for another small group of countries. These statistics were used together with emission factor information to assess trace metal emissions from waste incineration. The average emission factors are presented in Table 12. The results of emission estimates are given in Table 13. The statistical information was obtained from the UN Environment Programme, Environmental Data Report 1993–1994 (UNEP 1996), the OECD Environmental Data Compendium 1995 (OECD 1997), and the Statistical Compendium for the Dobris Assessment of Europe's Environment (EEA 1995).

Table 12. Emissions factor for waste disposal, in grams/tonne waste incinerated.

Elements	Municipal waste	Sewage sludge waste
As	1.1	5.0
Cd	0.4	1.0
Cr	0.7	50.0
Cu	7.0	10.0
Hg	1.0	5.0
Mn	1.8	50.0
Ni	0.7	10.0
Pb	10.0	80.0
Sb	3.0	5.0
Se	0.2	1.0
Sn	1.0	5.0
Zn	20.0	50.0
V		3.0

Table 13. Emissions of trace metals from (*a*) municipal waste, and (*b*) sewage sludge incineration in the mid-1990s (in tonnes/year).

Continent	As	Cd	Cr	Cu	Hg	Mn	Ni	Pb	Sb	Se	Sn	Zn	V
(*a*) *Municipal waste*													
Europe	29	10	18	182	10	47	18	250	78	5	26	520	
Asia	36	13	23	228	33	59	23	326	98	7	33	652	
N. America	21	8	14	136	64	35	14	194	58	4	19	388	
Australia and Oceania	1	1	—	1	—	—	—	1	1	—	—	3	
World total	87	32	55	547	107	141	55	771	235	16	78	1563	
(*b*) *Sewage sludge*													
Europe	3	1	26	5	1[a]	26	5	1[a]	3	1	3	26	2
Asia													
N. America	34	7	344	69	2	344	69	50	34	7	34	344	21
Australia and Oceania													
World total	37	8	370	74	2	370	74	50	37	8	37	370	23

[a] Emissions of Hg and Pb from sewage sludge incineration in Europe are included in the emissions of these metals from municipal waste incineration.

In general, emission data in Table 13 are very much incomplete and therefore largely underestimate, probably by one order of magnitude, for As, Cd, Cr, Pb, Sn, and Zn release. The incompleteness of data has been caused by a lack of information from most countries on emissions of trace metals during waste incineration and (or) on the amount of wastes incinerated. Once more reliable data on the magnitude of wastes incinerated and the content of trace metals in these wastes are obtained, additional focus should be put in future emission inventorying activities. This will result in more accurate and complete emission estimates.

4.3.6. Total worldwide emissions from anthropogenic sources

Results of estimates of worldwide emissions of trace metals from major anthropogenic sources to the atmosphere in the mid-1990s are presented in Table 14. These results indicate that stationary fossil fuel combustion continues to be the major source of Cr (69%), Hg (66%), Mn (85%), Sb (47%), Se (89%), Sn (89%), and Tl (almost 100%) with respect to coal combustion and the major source of Ni (90%) and V (almost 100%) with respect to oil combustion.

Combustion of leaded, low-leaded, and unleaded gasoline continues to be the major source of atmospheric Pb emissions, contributing about 74 % to the total anthropogenic emissions of this metal in 1995.

The third major source of trace metals is non-ferrous metal production, which is the largest source of atmospheric As (69%), Cd (73%), Cu (70%), In (100%), and Zn (72%).

It should be noted again that the information on the emissions of trace metals from waste incineration is clearly incomplete with respect to the number of countries reporting their emissions from this source and inaccurate with respect to the emission factors used to calculate trace metal emissions from waste incineration for countries not reporting their national emissions. In addition, statistical information on the amounts of municipal and sewage sludge wastes is also incomplete. The above-mentioned problems result in underestimation of trace metal emissions from this source, particularly for As, Cd, Cr, Mn, Pb, Sb, Se, Sn, and Zn.

Emissions of trace metals released mostly during non-ferrous metal production have decreased by a factor of 2 to 3 between the beginning of the 1980s and the mid-1990s, mostly because of the improvement of emission control efficiency in major smelters in Europe and North America. One cannot rule out another explanation for this change, that the emission factors used to estimate emissions at the beginning of the 1980s were somewhat overestimated for some metals, as already discussed by Skeaff and Dubreuil (1997).

Emissions of trace metals emitted mostly from coal combustion are lower by a factor of 2 in the 1990s compared to the emissions at the beginning of the 1980s, while emissions of V and Ni from oil combustion are higher by a factor of 2 to 3. This trend was explained earlier in this work.

Results of worldwide emission estimates of anthropogenic trace metals for various continents are presented in Table 15. Emissions from sources in Asia are clearly the largest for all metals estimated in this work. This can be related to the increase of industrial production in this part of the world. However, progress in combating environmental pollution does not always follow industrial growth, particularly in the Asian countries.

Concerning the contribution of individual countries to the total emissions of anthropogenic trace metals, this information has been presented in this work for Hg from stationary fossil fuel. The Asian and South American countries are also the largest emitters of As and Cd from the non-ferrous metal industry. High emissions are also estimated for the U.S. smelters. The above information is presented in Table 16. Unfortunately, no data exist on the emissions of trace metals from individual countries or even continents for the beginning of the 1980s. Therefore, the development of emission geography between the beginning of the 1980s and the mid-1990s is impossible to review.

There are no other data in the literature on emissions of several trace metals that could be compared with the estimates presented in this work. The only estimates of global anthropogenic emissions were found for Hg (Pirrone et al. 1996). A comparison of these estimates with the data presented in this work is shown in Table 17. Very good agreement was obtained for the total emissions and for Asia, the main region of anthropogenic Hg emissions to the atmosphere. Major differences were observed for Europe and Africa. Because most of the European emissions presented in this work are based on discussions with national emission experts in various European countries, therefore it is claimed that the data presented herein are more reliable.

Table 14. Worldwide emissions of trace metals from major anthropogenic source categories to the

Source category	As	Cd	Cr	Cu	Hg	In
Stationary fossil fuel combustion	809	691	10 145	7 081	1 475	
Vehicular traffic						
Non-ferrous metal production	3 457	2 171	—	18 071	164	45
Iron and steel production	353	64	2 825	142	29	—
Cement production	268	17	1 335	—	133	—
Waste disposal	124	40	425	621	109	—
Other					325[a]	
Total	5 011	2 983	14 730	25 915	2 235	45
1983 emission[b]	18 820	7 570	30 480	35 370	3 560	25

[a] Emission of Hg from gold production.
[b] Nriagu and Pacyna (1988).

5. Quality of emission estimates

It is very difficult to assess the accuracy of currently available emission data for trace metals mostly because of limited information on the accuracy of emission factor estimates and specific statistical data in various countries. Ideally, emissions from at least major sources, such as large power plants, waste incinerators, smelter, steel and iron plants, and cement kilns should be measured. The emission measurements would generate the most accurate data on emissions assuming that the proper sampling methods and analytical techniques are applied and the samples are collected at representative sites along the path of flue gases in the stack. However, to measure emissions at so many point sources of emissions is for many reasons impossible to accomplish in very many countries. Therefore, less accurate methods based on emission factors and material balances must be applied.

The information for Pb is claimed to be the most accurate, as most Pb emissions on a global scale originate from combustion of leaded gasoline. Information on consumption of gasoline in various countries is quite accurate, as is information on the use of lead additives. A comparison of various independent estimates for Europe as a whole and for individual countries seems to indicate an accuracy of about 15%.

Cadmium and mercury emitted from anthropogenic sources have been the subject of several emission estimates. Their major sources are fairly well defined and emission factors established. An emission estimation accuracy of better than 50% can be assigned for these two trace metals in Europe, as proved by the comparison of air concentrations calculated by models using emission data and those measured. A very recent study carried out at the Meteorological Synthesizing Centre-East (MSC-E) of EMEP (EMEP MSC-E) concluded that an agreement between the model estimates based on the emission data for Europe, used in this paper, and air measurements at various EMEP stations in Europe was as good as 6% for Pb, 6% for Hg, and 14% for Cd (Ryaboshapko et al. 1999). No analysis was carried out for other trace metals. These results indirectly confirm very good accuracy of emission estimates presented in this paper, at least for Europe. They also indicate a proper selection of emission factors for emission estimates presented here.

Similar comparison made by other research groups (e.g., Olendrzynski et al. 1996) for Zn seems to indicate an accuracy within a factor of 2. In general, the Zn data are often underestimated due to incomplete knowledge of emission sources. While emissions of Zn from zinc non-ferrous metal industry are quite accurate, there are many sources related to the Zn consumption, and the Zn emissions from these sources are often less accurately estimated.

In summary, only single estimates of emissions are presented in this work for the studied trace metals

Table 14. atmosphere in the mid-1990s (in tonnes/year).

Mn	Mo	Ni	Pb	Sb	Se	Sn	Tl	V	Zn
9 417	2 642	86 110	11 690 88 739	730	4 101	3 517	1 824	240 084	9 417
59	—	8 878	14 815	552	466	319	—	77	40 872
1 060	—	36	2 926	7	7	—	—	71	2 118
—	—	134	268	—	3	—	—	—	2 670
511	—	129	821	272	24	115	—	23	1 933
11 047	2 642	95 287	119 259	1 561	4 601	3 951	1 824	240 255	57 010
38 270	3 270	55 650	332 350	3 510	3 510	3 790	5 140	86 000	131 880

and source categories. In the case that emissions were estimated using emission factors, these factors were the ones selected as the most relevant for a given industrial technology, an industrial development in a given country or region, progress and improvements in application of advanced emission control equipment, the content of trace metals in raw materials, etc. However, emission estimates for one category can be more accurate and complete than the estimates for another source category. It is concluded here that the following accuracy of emission estimates can be assigned to the estimates for individual source categories in this work: stationary fossil fuel combustion, ±25%; combustion of gasoline, ±15%; non-ferrous metal production, ±20%; iron and steel production, ±30%; cement production, ±30%; and waste disposal, a factor of up to 5.

The most accurate emission estimates are those calculated for gasoline combustion. Leaded, low-leaded, and unleaded gasoline consumption in various countries is known within 15%, and this can also be accepted for worldwide Pb emission estimates. Most of the emission factors used in the project for fossil fuel combustion, cement production, and pig iron and steel production (Table 2) are within the ranges of emission factors presented in the UN ECE Guidebook (UN ECE 1999*b*) and differ from the emission factors defined in this guidebook as suggested for use within the above-mentioned percentage. The same applies to the emission factors for non-ferrous metal industry (Table 9).

Emission factors for waste disposal, available from the above-mentioned guidebook, differ by a factor of up to 5 compared to the data in Table 12. Clearly, the trace metal emission estimates for waste disposal are the least accurate compared to estimates for other source categories considered in this work.

The above-mentioned values apply to global estimates. As the emission data for several countries in Europe and North America have been evaluated by national emission experts, it can be suggested that the emission estimates for Europe and North America may be more accurate than the emission estimates for other continents.

The strengths of current emission estimates are found in improved completeness of emission source categories considered and in the transparency of emission data presentation. Whereas more accurate emission data for trace metals are becoming available for major source categories, including the combustion of fuels, the non-ferrous metal production, and the production of certain industrial goods, some fresh information on the amounts and emission factors now exists for other sources, including various applications of trace metals as additives, etc. Improvements with respect to the completeness of emission data for major source categories are due to emission measurements often carried out in connection with various environmental protection regulations being introduced more frequently at present, compared to the past 10–20 years. These regulations can be national or international, e.g., the agreements on emission

Table 15. Worldwide emissions of trace metals from major anthropogenic source categories to the atmosphere in the mid-1990s, on the continent by continent basis (in tonnes/year).

Continent	As	Cd	Cr	Cu	Hg	In	Mn	Mo	Ni	Pb	Sb	Se	Sn	Tl	V	Zn
Europe	607	362	3 353	2 245	313	8	2 339	531	20 417	28 091	273	838	863	311	57 144	7 689
Africa	324	172	847	2 031	389	2	738	207	10 690	11 349	66	300	425	78	28 930	2 353
Asia	2416	1463	6 234	12 979	1121	20	4 482	1115	41 228	51 212	694	1982	1676	771	101 314	34 886
N. America	658	482	3 284	2 841	215	6	2 670	579	11 236	17 015	375	1086	531	564	26 660	5 859
S. America	925	452	623	5 453	84	8	497	128	11 092	9 118	101	225	426	1	25 443	5 353
Australia and Oceania	81	52	389	366	113	1	321	82	624	2 474	52	170	30	99	764	870
Total	5011	2983	14 730	25 915	2235	45	11 047	2642	95 287	119 259	1561	4601	3951	1824	240 255	57 010

Table 16. The 10 largest emitter countries for As and Cd from non-ferrous metal industry in 1995 (emissions in tonnes).

	As		Cd	
No.	Country	Emission	Country	Emission
1	Chile	644	China	440
2	China	600	Japan	356
3	Japan	591	Chile	258
4	United States	200	United States	112
5	Zambia	157	Korea	104
6	Peru	150	Peru	89
7	Kazakhstan	136	Kazakhstan	82
8	Korea	131	Brazil	71
9	Brazil	92	Zambia	63
10	Philippines	79	Canada	61

Table 17. Comparison of estimates of worldwide anthropogenic Hg emissions to the atmosphere by region, reported for 1992 by Pirrone et al. (1996), and this work for 1995 (in tonnes/year).

Continent	Pirrone et al. (1996) for 1992	This work for 1995
Europe	634	313
Africa	113	389
Asia	1012	1121
North America	332	215
South America	73	84
Australia and Oceania	35	113
Total	2199	2235

reductions for trace metals, recently established within the UN ECE. The European Union is also in the process of preparing directives aimed at the reduction of Hg and other trace metals in the EU region.

In connection with international agreements on the reduction of emissions of trace metals to the air and their loads to the marine and terrestrial ecosystems, countries in various regions, such as Europe are obligated to report their emission data to the international organizations responsible for the implementation of the results of these agreements. This reporting is required in a very transparent manner, which has a direct impact on the improvement of transparency of emission estimates. Guidebooks and guidelines are now becoming available within the international and national organizations, advising emission experts on how to estimate and report emissions of trace metals. An example could be a Joint EMEP/CORINAIR Atmospheric Emissions Inventory Guidebook (UN ECE 1999*b*). This Guidebook is quite extensively used, which contribute to the improvement of completeness, accuracy, and transparency of emission data for trace metals being generated at present, mostly in Europe and North America, but also in Asia, South America, and recently in Africa.

However, these positive developments in collating of accurate emission data for trace metals from anthropogenic sources have not yet overcome the shortcomings in emission inventorying related mainly to still weak communication links between emission experts preparing inventories at a national level and industry and other environmental change drivers. The developments within this work have been an important step towards the improvement of this communication.

Table 18. A comparison of estimated global anthropogenic emissions of trace metals in the mid-1990s with emissions from natural sources (Nriagu 1989) (emissions in 10^3 tonnes/year).

Trace metal	Anthropogenic emissions	Natural emissions: median values	Anthropogenic/national emission ratios
As	5.0	12.0	0.42
Cd	3.0	1.3	2.3
Cr	14.7	44.0	0.33
Cu	25.9	28.0	0.93
Hg	2.2	2.5	0.88
Mn	11.0	317.0	0.03
Mo	2.6	3.0	0.87
Ni	95.3	30.0	3.2
Pb	119.3	12.0	9.9
Sb	1.6	2.4	0.67
Se	4.6	9.3	0.49
V	240.0	28.0	8.6
Zn	57.0	45.0	1.3

6. Comparison of contributions of emissions from natural and anthropogenic sources to the total global emission of trace metals

Estimation of trace metal emissions from natural sources was not within the scope of this work. A new study on the estimates of trace metal emissions from natural sources is now being carried out at the Geological Survey of Canada.[3] The results of this study are expected in the near future. However, at least some comparison of the estimates of trace metal emissions from anthropogenic sources, presented in this work with the older estimates for natural sources available from the literature, can be approached. An important contribution to describing the significance of natural sources of trace metals in the environment is that by the Geological Survey of Canada (GSC 1995), but the widest quantitative assessment of emissions from these sources is still the one by Nriagu (1989). Obviously, there are also other emission estimates, but they are mainly limited to a certain trace metal or to a single emission source category.

The GSC report discusses releases of trace metals from deflated soil and sediment, volcanic emissions, forest fire debris, biogenic emissions, and oceanic emissions. General conclusion has been reached that there are significant uncertainties in comparing natural and anthropogenic emissions of trace metals, particularly on a global scale. Natural emission estimates vary widely, since they are extrapolated from very sparse data sets. In such cases the estimated ratios of natural and anthropogenic emissions are uncertain.

A comparison of global anthropogenic emission estimates in this work with the global natural emission estimates by Nriagu (1989) is presented in Table 18. This comparison suggests that anthropogenic emissions of Pb and V are by one order of magnitude higher than the natural emissions of the these metals on a global scale. Anthropogenic emissions are a factor of 2 to 3 higher than the natural emissions for Cd and Ni, and they are comparable for Cu, Hg, Mo, Sb, and Zn. The comparison in Table 18 also indicates that global natural emissions of As, Cr, and Se are larger than the global anthropogenic emissions of these elements by a factor of 2 to 3. Finally, the natural sources are by far more significant than the anthropogenic sources for Mn.

[3] Robert Garrett, personal communication

There are a few interesting observations that can be made on the basis of the comparison in Table 18. In general, there are no major surprises in this comparison for the mid-1990s compared to our previous findings with respect to the significance of anthropogenic sources of trace metals to the total budget of these contaminants. Perhaps the only exception is for As, often regarded as very much an anthropogenic trace metal. One should add that the As emission from natural sources considered in Table 18 is quite high and probably overestimated. For Hg, anthropogenic sources seem to be as important as natural sources. It should be underlined that the suggestions presented above are valid when we discuss the significance of anthropogenic versus natural sources on a global scale. This significance can be entirely different when discussing the situation on a local scale, e.g., around a waste incinerator or a volcano and even on a regional scale, e.g., in Central Europe or in the Mediterranean Sea basin.

7. Final remarks

An accurate and complete emission inventory for atmospheric trace metals on a global scale is needed for both modeler community and policy makers to assess the current level of environmental contamination by these pollutants, major emission sources and source regions, and the contribution of atmospheric pathway to the contamination of terrestrial and aquatic environment. International policy makers would need the information on global emissions to properly analyze source contribution to the pollution of a certain region and then to propose proper abatement strategy. Modelers would need the emission survey to provide with the scientific substantiation material for these strategies through the application of global models of trace metal transport within air masses. Major progress has been made in assessing emissions of trace metals in various countries and even regions, e.g., Europe since the first global emission estimate for these pollutants was made by Nriagu and Pacyna (1988). These improved national and regional emission inventories have been used in this work to assess the global trace metal emissions from anthropogenic sources in the mid-1990s.

The results of this work conclude that stationary fossil fuel combustion continues to be the major source of Cr, Hg, Mn, Sb, Se, Sn, and Tl with respect to coal combustion and the major source of Ni and V with respect to oil combustion. Combustion of leaded, low-leaded, and unleaded gasoline continues to be the major source of atmospheric Pb emissions. The third major source of trace metals is non-ferrous metal production, which is the largest source of atmospheric As, Cd, Cu, In, and Zn.

The largest anthropogenic emissions of atmospheric trace metals were estimated in Asia. This can be explained by growing demands for energy in the region and increasing industrial production. As a result, the Asian emissions are not only larger than the emissions on other continents, but also show an increasing trend. Another factor contributing to high emissions in Asia is the efficiency of emission control, which is lower than in Europe and North America. Concerning the two latter continents, emissions of trace metals show a decreasing tendency over the last two decades.

The emission inventory presented in this work is based on emission estimates by both national experts in several countries in Europe and North America and author estimates for the remaining countries. National estimates are sometimes based on emission measurements (in non-ferrous metal smelters) that are considered a reliable source of information when preparing emission inventories. Indeed, more emission measurements should be carried out to further improve emission inventories. Emission measurements should be suggested for at least major point sources of emissions, such as power plants, smelters, waste incinerators, and cement kilns.

Most of the trace metals are emitted on fine particles, while a portion of Hg and Se is emitted in a gas phase. Entering the atmosphere on fine particles and in a gaseous form, trace metals can be transported with air masses on long distances, e.g., 2000 km. There is fairly solid scientific evidence of such transport. It is very important to be able to conclude to what extent concentrations measured in remote locations are of anthropogenic origin and what is the contribution of emissions from natural sources. Such information is crucial for preparing proper strategies on emission reduction with respect to

emissions from anthropogenic sources. The quality of this information is directly related to the accuracy and completeness of emission inventories for both anthropogenic and natural sources.

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References

Axenfeld, F., Munch, J., Pacyna, J.M., Duiser, J.A., and Veldt, C. 1992. Test-Emissionsdatenbasis der Spurenelemente As, Cd, Hg, Pb, Zn und der speziellen Organischen Verbindungen y-HCH (Lindan), HCB, PCB und PAK fur Modellrechnungen in Europa. Umweltforschunsplan des Bundesministers fur Umwelt Naturschutz und Reaktorsicherheit, Luftreinhaltung, Forschungsbericht 104 02 588, Dornier GmbH Report, Friedrichshafen, Germany.

Arctic Monitoring and Assessment Programme (AMAP). 1997. AMAP assessment report: Arctic pollution issues. Arctic Monitoring and Assessment Programme, Oslo, Norway.

Arctic Monitoring and Assessment Programme (AMAP). 1999. Heavy metals. Chapter 7. AMAP assessment report: Arctic pollution issues. Arctic Monitoring and Assessment Programme, Oslo, Norway.

Berdowski, J.J.M., Pacyna, J.M., and Veldt, C. 1994. Emissions. *In* The impact of atmospheric deposition of non-acidifying pollutants on the quality of European forest soils and the North Sea. *Edited by* K.D. van den Hout, Air and Energy Directorate of the Dutch Ministry of Housing, Spatial Planning and the Environment (VROM), The Hague, The Netherlands.

Berdowski, J.J.M., Baas, J., Bloos, J.P.J., Visschedijk, A.J.H., and Zandveld, P.Y.J. 1997. The European emission inventory of heavy metals and persistent organic pollutants. Umweltforschungsplan des Bundesministers fur Umwelt, naturschutz und Reaktorsicherheit. Forschungsbericht 104 02 672/03, Apeldoorn, the Netherlands, June.

Boyd, R., Barnes, S.-J., De Caritat, P., Chekustin, V.A., Melezhik, V.A., Reimann, C., and Zientek, M.A. 1998. Assessment of heavy metal emissions from the copper–nickel industry on the Kola Peninsula and other parts of Russia. NGU Report 98.138, Geological Survey of Norway, Trondheim, Norway, November.

European Environment Agency (EEA). 1995. Statistical compendium for the Dobris assessment of Europe's environment. European Environment Agency (EEA), Copenhagen, Denmark.

Gromov, S.A. 1996. An estimate of lead emissions from road transport at the USSR territory in 1990. *In* Report and Proceedings of the WMO Workshop on the Assessment of EMEP Activities Concerning Heavy Metals and Persistent Organic Pollutants and Their Further Development. EMEP/ MSC-E report 1/97, Moscow, Russia. pp. 120–124.

Geological Survey of Canada (GSC). 1995. The significance of natural sources of metals in the environment. *Prepared by* the Geological Survey of Canada for the UN-ECE LRTAP (Heavy Metals) Convention, Natural Resources Canada, Ottawa, Ont.

ITIT. 1992. Proceedings of the ITIT International Symposium on Emission Inventory and Prevention Technology for Atmospheric Environment, Tsukuba, Japan.

Jacques, A.P. 1987. Summary of emissions of antimony, arsenic, cadmium, copper, lead, manganese, mercury, and nickel in Canada. Environmental Analysis Branch, Conservation and Protection, Environment Canada, Ottawa, Ont.

Lacerda, L.D. 1997. Global mercury emissions from gold and silver mining. Water, Air, Soil Pollut. **97**: 209–221.

Norwegian Institute for Air Research (NILU). 1984. Emission sources in the Soviet Union. NILU TR 4/84 Report. The Norwegian Institute for Air Research, Kjeller, Norway.

Nriagu, J.O. 1989. A global assessment of natural sources of atmospheric trace metals, Nature, **338**: 47–49.

Nriagu, J.O., and Davidson, C.I. 1986. Toxic metals in the atmosphere. John Wiley and Sons, New York, N.Y.

Nriagu, J.O., and Pacyna, J.M. 1988. Quantitative assessment of worldwide contamination of air, water and soils with trace metals. Nature, **333**: 134–139.

Nriagu, J.O., Blankson, M.L., and Ocran, K. 1996. Childhood lead poisoning in Africa: a growing public health problem. Sci. Total Environ. **181**: 93–100.

Organisation for Economic Co-operation and Development (OECD). 1997. Environmental data compendium 1995. Organization for Economic Co-operation and Development, Paris, France.

Olendrzynski, K., Anderberg, S., Bartnicki, J., Pacyna, J.M., and Stigliani, W. 1996. Atmospheric emissions and depositions of cadmium, lead and zinc in Europe during the period 1955–1987. Environ. Rev. **4**: 300–320.

Pacyna, J.M. 1984. Estimation of the atmospheric emissions of trace elements from anthropogenic sources in Europe. Atmos. Environ. **18**: 41–50.

Pacyna, J.M. 1994. Long-range transport, atmospheric dispersion and deposition. *In* Heavy metals emissions, UN ECE Task Force on Heavy Metals Emissions State-of-the-Art Report, Prague, the Czech Republic.

Pacyna, J.M. 1996. Atmospheric emissions of heavy metals for Europe. Improvements, updates, historical data and projections. A Report for the International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria, June.

Pacyna, J.M. 1998. An assessment of the applicability of the critical load concept as a basis for international protocols on emission reductions for heavy metals and persistent organic compounds. Sources, transport and deposition. *In* Assessment of the applicability for pollution control authorities of the concept "critical load" of long-range transported micropollutants in relation to aquatic and terrestrial ecosystems. Research report no. 32 of the Ministry of Environment, Oslo, Norway. pp. 31–40.

Pacyna, J.M., and Pacyna, E.G. 1999. Atmospheric emissions of anthropogenic lead in Europe: improvements, updates, historical data and projections. Technical Report for GKSS Research Center, Hagan, Norway, June.

Pacyna, J.M., and Pacyna, P.E. 1996. Global emissions of mercury to the atmosphere. Emissions from anthropogenic sources. *Prepared for* Report for the Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. June.

Pacyna, J.M., Scholtz, M.T., and Li, Y.-F. 1995. Global budget of metal sources. Environ. Rev. **2**: 145–159.

Pirrone, N., Keeler, G.J., and Nriagu, J.O. 1996. Regional differences in worldwide emissions of mercury to the atmosphere. Atmos. Environ. **30**: 2981–2987.

Porcella, D.B., Chu, P., and Allan, M.A. 1996. Inventory of North American Hg emissions to the atmosphere. *In* Global and regional mercury cycles: Sources, fluxes and mass balances. *Edited by* W. Baeyens, R. Ebinghaus, and O. Vasiliev. NATO ASI Series, Series 2: Environment — Vol. 21, Kluwer Academic Publishers, Dordrecht, The Netherlands.

Ryaboshapko, A., Ilyin, I., Gusev, A., Berg, T., and Hjellbrekke, A.-G. 1999. Monitoring and modelling of lead, cadmium and mercury transboundary transport in the atmosphere of Europe. EMEP Report 3/99. The UN ECE Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). Meteorological Synthesizing Centre — East, Moscow and Chemical Coordinating Centre, Kjeller, Norway.

Skeaff, J.M., and Dubreuil, A.A. 1997. Calculated 1993 emission factors of trace metals for Canadian non-ferrous smelters. Atmos. Environ. **31**: 1449–1457.

Tsibulski, V., Yatsenko-Khemlevskaya, M., and Milyaev, V. 1996. Estimation of heavy metal emissions due to organic fuel combustion in Russia in 1990 and 1993. *In* Report and Proceedings of the WMO Workshop on the Assessment of EMEP Activities Concerning Heavy Metals and Persistent Organic Pollutants and Their Further Development. EMEP/MSC-E report 1/97, Moscow, Russia. pp. 115–119.

United Nations (UN). 1997. Statistical Yearbook, United Nations, New York, N.Y.

United Nations (UN). 1998. Industrial Commodity Statistics Yearbook 1996. United Nations, New York, N.Y.

United Nations Economic Commission for Europe (UN ECE). 1994. Heavy metals emissions, Task Force on Heavy Metals Emissions State-of-the-Art Report, Prague, The Czech Republic.

United Nations Economic Commission for Europe (UN ECE). 1998. Air pollution. The convention on

long-range transboundary air pollution. International cooperation in action. The United Nations Economic Commission for Europe, Geneva, Switzerland.

United Nations Economic Commission for Europe (UN ECE). 1999*a*. EMEP emission data. EMEP/MSC-W Report 1/99. The UN ECE Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). Meteorological Synthesizing Centre–West, Oslo, Norway.

United Nations Economic Commission for Europe (UN ECE). 1999*b*. Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook. The United Nations Economic Commission for Europe, Geneva, Switzerland.

United Nations Environment Programme (UNEP). 1996. Environmental Data Report 1993–1994. United Nations Environment Programme, Nairobi, Kenya.

United States Environmental Protection Agency (US EPA). 1993*a*. Locating and estimating air emissions from sources of mercury and mercury compounds. EPA Rept. 454/R-93–023, United States Environmental Protection Agency, Research Triangle Park, N.C.

United States Environmental Protection Agency (US EPA). 1993*b*. Locating and estimating air emissions from sources of cadmium and cadmium compounds. EPA Rept. 454/R-93–040, United States Environmental Protection Agency, Research Triangle Park, N.C.

World Bureau of Metal Statistics (WBMS). 1999. World metal statistics. World Bureau of Metal Statistics, May.

Appendix A. A list of reports with national emission data for trace metals

1. Austria

   Abschatzung der Schwermetallemissionen in Østerreich, Umweltbundesamt, UBE Rept.-95-108, Wien, 1995, and

   Winiwarter, W. Emission of Cd. Hg and Pb in Austria in 1985, 1990, and 1995. Austrian Research Centers, Seibersdorf, December 10, 1998.

2. Belarus

   Kakareka, S., Estimation of lead emissions for Belarus in SNAP – source category. Belarusian National Academy of Sciences, Institute for Problems of Use of Natural Resources and Ecology, Minsk, December 30, 1998, and

   Belarusian Contribution to EMEP, Annual Report, 1996. Belarusian National Academy of Sciences, Meteorological Synthesizing Center – East, Minsk – Moscow, January 1997.

3. Bulgaria

   Raykova, S. The Elbe catchment to Pb emission scenarios. Ministry of Environment and Water, Sofia, January 1, 1999, and

   Syrakov, D. 1997 Annual Report on Bulgarian Contribution to EMEP. National Institute of Meteorology and Hydrology, Sofia, January 30, 1998.

4. The Czech Republic

   Data reported to the UN ECE Task Force on Heavy Metals,

   Machalek, P. Emissions estimates data –HMs in Czech Republic (1990–1995). Czech Hydrometeorological Institute, Emission Inventory Section, Division of Air Quality Protection, Prague, January 28, 1999, and

   CHI: Air Pollution in the Czech Republic in 1997. The Czech Hydrometeorological Institute (CHI), Air Quality Protection Department, Prague, 1998.

5. Denmark

Data reported to the UN ECE Task Force on Heavy Metals, and to the PARCOM/ATMOS program, and

Winther, M. The Danish Pb air emissions from the Danish CORINAIR 1997 inventory. Ministry of Environment and Energy, National Environmental Research Institute. Department of Policy Analysis, Copenhagen, January 12, 1999 and January 28, 1999.

6. Finland

Data reported to the UN ECE Task Force on Heavy Metals, and

Mukherjee, A.B. Emissions of Pb in Finland. University of Helsinki, Department of Limnology and Environmental Protection, Helsinki, December 1998.

Mukherjee, A.B. The occurrence of nickel in the Finnish environment. University of Helsinki, Department of Limnology and Environmental protection, Helsinki, 1998.

7. France

Fontelle, J.P. Heavy Metals emissions based on the 11 CORINAIR/MNECE/EMEP groups (take care of version SNAP 1997). CITEPA (Centre Interprofessionnel Technique d'ÉTUDES de la Pollution Atmospherique), Paris, February 2, 1999.

8. Germany

Schwermetallemissionen in die Atmosphare, TUV Rheinland, Koln, EP 10/93, 1993, and

Jockel, W., Hartje, J. Report on the Development of the Emissions of Heavy Metals in Germany 1985–1995. Umweltforschungsplan Des Bundesministers für UMWELT, Naturschutz und Reaktorsicherheit, Köln, July 1997, letter of Jockel, W. Environmental Protection/Air Pollution Control, Köln, December 2, 1998.

9. Hungary

Kozak, K. Emissions of Pb in Hungary, 1980–1997. Ministry of Environment Republic of Hungary, Department of Environmental Development, Safety and Waste Management, Budapest, January 8, 1999.

10. Italy

Costa, P. Emission of Pb. CNR-Istituto Sull'Inquinamento Atmosferico, IIA-CNR, Universita della Calabria, Rende, December 3, 1998.

11. Lithuania

Semlstoriute, D. Pb emissions in Lithuania. Ministry of the Environment, Environmental Quality Department, Air Division, Vilnius, December 16, 1998, and

Brazauskas, R. Pb emissions in Lithuania. Ministry of the Environment, Air Division of the Environmental Quality Department, Vilnius, January 19, 1999.

12. The Netherlands

Emission inventory in the Netherlands. Emissions to air and water in 1992. The Ministry of Housing, Spatial Planning and the Environment, Directorate-General for the Environment, Gravenhage, Publ. No. 22, December 1994, and

Berdowski, J.J.M. et al. Emissions to air for the Inventories of CORINAIR, EMEP and OSPARCOM 1990–1996. Ministry of Housing, Spatial Planning and the Environment, January 1998.

13. Norway

Miljøgifter i Norge. Statensforurensningstilsyn (SFT), Rept. 92/103, Oslo, 1993,

14. Poland

Report on Heavy Metals Emissions in Poland for 1990. The Institute for Ecology of Industrial Areas, Katowice, November 1993, and

Mitosek, G. National annual total antropogenic emissions of heavy metals 1990–1995. Institute of Environmental Protection, Air Protection Division, Warsaw, October 1997, and

Hlawiczka, S. Report on heavy metals emission in Poland for the year 1996. Institute for Ecology of Industrial Areas, Katowice, March 1998.

15. Slovenia

Rode, B. Heavy Metals emission estimations of Cd, Hg and Pb based on Technical Paper to the OSPARCOM-HELCOM UNECE Emission Inventory, 1990–1996. Ministry of the Environment and Physical Planning Hydrometeorological Institute of Slovenia, Ljubljane, December 29, 1998.

16. Slovakia

Heavy Metals in Slovakia. Ministerstvo Zivotneho Prostredia Slovenskej Republiky, Bratislava, December 1994.

17. Sweden

Heavy Metal Emissions to Air in Sweden in 1992. Swedish Environmental Protection Agency, Industrial Department, Stockholm, December 1993.

18. Switzerland

Vom Menschen Verursachte Schadstoff-Emissionen in der Schweitz 1950–2010. Bundesamt fur Umweltschutz, Schriftenreiche Umweltschutz No. 76, Bern, and

Saxer, H.P. “Schriftenreihe Umwelt Nr. 256” of BUWAL, Emissions of Lead in Switzerland. 1950–2010. Swiss Agency for the Environment, Forests and Landscape, Section Biotechnology and Flux of Substances, Berne, January 5, 1999.

19. United Kingdom

The U.K. Atmospheric Emissions of Metals and Halides 1970–1991. National Atmospheric Emissions Inventory. Department of Environment, London, September 1993.

20. United States

National Air Pollutant Emission Trends 1900–1995. The U.S. Environmental Protection Agency, US EPA-454/R-96-007 Report, Research Triangle Park, N.C., 1996.