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ANTHROPOGENIC AIR POLLUTION IN THE ANCIENT TIMES

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Összefoglalás – Napjainkig számos összefoglaló kötet, illetve tanulmány jelent meg az elmúlt korok környezetszennyezéséről (pl. *Brimblecombe*, 1987; *Brimblecombe and Pfister*, 1990; *McNeill*, 2001, *Bowler and Brimblecombe*, 2000; *Karatzas*, 2000, 2001). E dolgozat célja, hogy további adalékokkal szolgáljon a tárgykörben – elsősorban az ókorból és a középkorból.

Summary – Several comprehensive publications have been issued recently on the environmental pollution of the past times (e.g. *Brimblecombe*, 1987; *Brimblecombe* and *Pfister*, 1990; *McNeill*, 2001, *Bowler and Brimblecombe*, 2000; *Karatzas*, 2000, 2001). The aim of the study is to give further information on the subject – mainly in the ancient and the medieval times.

Key words: lead mining, copper mining, lead pollution of teeth, lead pollution of the atmosphere, copper pollution of the atmosphere

POLLUTION OF THE ENVIRONMENT IN THE ANCIENT TIMES

Pollution of the environment has started with the appearance of humans. When Homo Sapiens lighted fire, its smoke proved to be the first environmental pollution. Air pollution of inner spaces has started with using fuels for heating and cooking. Walls of caves, inhabited many thousand years ago, are covered by thick layers of soot. Hence, it is supposed that breathing of cavemen was difficult due to the smoke, which also irritated their eyes in the closed room. Lungs of mummified bodies from the Palaeolithic era are frequently black. In the first places, which served for living, smoke was not driven away (one of its practical reasons might have been protection against mosquitoes) and cavemen then lived in smoky rooms (McNeill, 2001). [Millions of people have recently been living in this way. In 1993, when we were in Nepal, we were trekking in the Langtang National Park and visited many little villages and stayed at "hotels", when going on the southern slopes towards the High Himalayas. Even recently, smoke of fire is not driven away from buildings here. Walls of the houses are built of metamorphic slates, without binder and their roof are covered with rush matting. When lighting fire, there is thick smoke inside the house, irritating the eyes and making difficult breathing. It is impossible to sleep, even one can only stay there for a short time. And from outside, the house seems to be on fire, smoke is streaming out through the slits and gaps of the walls.] One has been living together with this harmful effect of air pollution for many thousand years.

Pollution of the environment was responsible for several kinds of illnesses in the early times. The very first pollution of the environment might have been the human excrement. Bowel bacterium living in the human body, such as the Escherichia coli, might have got from faeces to springs, which might have infected the early humans. This environmental pollution has been the reason of millions' illnesses even recently. In China, where a comprehensive system was developed for waste salvage even in the ancient times, fertilization with faeces was an important element of the agriculture even many thousand years ago. Productivity of the alluvial plain in the eastern part of the country has been maintained in this way for over 4,000 years. In several regions of China this tradition has been followed even recently. Han Suyin took the following note: "In Chengtu (capital of Sechuan Province) those families, who owned the city channel and, in this way, could send the accumulated faeces in countryside, have belonged to the richest ones even in the 20th century (till 1949)" (Markham, 1994). Fertilization of rice-paddies with faeces contributed largely to pollution of ground water which, in this way, is unfit for drinking in the whole tropic Asia. If, on the other hand, water is boiled, then salts are deposited and, in this way, it loses its taste. Flavouring boiled water with tee leaf comes from China and it started to spread in the empire about 2,000 B.C. and then all over Asia (Makra, 2000).

Dust pollution also appeared in the early times. According to the assumption of Janssens, in the New Stone Age in stone mines, e.g. in Obourg, people who carved flint from limestone day by day might have suffered from silicosis. The reason of it was that they breathed stone powder all the day. Sometimes the geographical position of the place considered was the cause of appearance of some diseases. Investigations revealed that near Broken, in the territory of the recent Zambia, Hominides who lived about 200,000 years ago, suffered from lead poisoning. The reason of this illness was that lead oozed from the neighbouring seam into the spring near their cave (*Markham*, 1994).

Effect of damaging the environment by ancient civilizations caused long-lasting changes in the environment, which can be experienced even recently. These effects appeared on regional scale; however, they did not cause global changes. During the 1,700 years long period between 3,500 B.C. and 1,800 B.C. in the plain of Tiger and Euphrates Rivers, efficiency of the Sumerian agriculture worsened more and more and wheat production decreased gradually, because the soil became salty. Water used for irrigation raises ground water level, and if redundant water is not driven away by channels, then soil is saturated with water, salts loose from soil and are deposited on the surface and then make an unbroken layer. Sumerian noted this process as "the soil surface became white". Water used for irrigation washed soils totally and made the region more and more unfit for agricultural production. This phenomenon largely contributed to the decline of the Sumerian culture. (*Markham*, 1994; *Mészáros*, 2002).

In the ancient times air pollution had substantial consequences only in cities. In the early towns, as in some recent settlements, penetrating stink might have frequently feeled, source of which was tainted meat, rotten foods and excrement. In these settlements, during siege – when there was no possibility to remove materials emitting aggressive smells – unbearable circumstances were developed. Egyptian historical records mention that when Nubian besieging troops cut off Hermopolis – which is situated on the left bank of the Nile, half-way between Theba and Memphis – the inhabitants rather handed over the town and presented a petition for peace than to bear further their own stinking air (*Brimblecombe*, 1995). In the ancient cities smell pollution was generally important. Aristoteles (384 B.C. – 322 B.C.) mentioned in his work *Athenaion Politeia* a rule that mature should be placed outside the town, at least 2 km away from the town walls (*Mészáros*, 2001). Smoke made

marble grey in antique towns, which annoyed classic poets as well [e.g. Horatius (65 B.C. – 8 A.D.)] and made, among others, ancient Jews to introduce list of laws (*Mamane*, 1987). In the ancient times air pollution was represented by smoke and soot.

There are several examples for polluting the environment in China, too. Before the Tang era (618-907), pine tree of the mountains in Shantung were burned, then in the Tang era Taihang mountains became bare in the border of Shansi and Hopei provinces (*Schäfer*, 1962). Similarly, during the Tang dynasty forests were cut around Loyang, the capital, in a circle with a radius of 200 miles. Trunk of the trees was burned in order to get ink for the governmental offices (*Epstein*, 1992).

Urban air pollution depends on the dimension of the given settlement, on the extension of the built-up territory as well as on the nature of the industrial activity, especially on using of traditional fuels. As urbanization has progressed in China, in the Mediterranean Basin and in North-western Africa, from about 1000 A.D, more and more people lived in smoky and sooty surroundings. Maimonides, the philosopher and physicist (1135-1204), who had comprehensive experiences on the towns of that era from Cordoba to Cairo, found that urban air is "stuffy, smoky, polluted, obscure and foggy", furthermore he thought that this condition is produced by "dullness preventing understanding, lack of intelligence and amnesia" being in the inhabitants (*Turco*, 1997).

On the other hand, traffic difficulties restricted air pollution of cities. Industrial activities consuming the most energy (e.g. tiles, glass, pottery, bricks and iron) were located near forests, since transporting mass of fuels to cities would have been too expensive. In this way, though air pollutants of industrial origin made air smelling bad, few people breathed it. Port cities were partially exceptions, as ships could transport wood and charcoal cheaply. Hence, Venice could maintain its glass industry, energy supply of which was arranged by transporting wood from far away. However, most part of urban air pollution came from household fuels, such as mature or wood but sometimes smokeless charcoal (McNeill, 2001). The air of the Chinese cities might have been extremely polluted, too, since the developed water transport system (Big Channel) permitted using high quantity of fuel, at least in the Sung capital, Kaifeng. Kaifeng (500 km south of Peking) was probably the first city in the world, which converted its energy supply from wood to coal. The transition was performed at the end of the 11th century, when the city had about one million inhabitants. However, coal heating period was short, because Mongolian troops destroyed Kaifeng in 1126 and those who remained in the city died from plague in the early 13th century (Hartwell, 1967).

Heavy pollution of the environment appeared simultaneously with developing of societies. Extensive environmental losses occurred even in the earliest societies. Air and water were polluted, soils were destroyed, species of plants and animals were extirpated. However, environmental changes made by the earliest societies, were little – the environment regenerated soon. Due to this, many people do not know anything about environmental losses of the early societies and, consequently, they are indulgent with early humans comparing with modern men living in urban environment. At the same time, there are examples of such environmental activities in ancient times, which made changes lasting even recently. Cutting down of forests in large areas for building ships in the ancient times might have contributed to decrease of forests' ratio in the Balkan Peninsula and in the territory of Greece. However, it is also possible that extensive destruction of forests occurred due to the drier summers in the Mediterranean (*Karatzas, 2000*). Nevertheless, this latter fact has no any relation with human activities. In Greece, due to little summer precipitation, stunted plants and bushes develop, which ensure grazing of ship and goat

having least demand. These animals, overgrazing slopes of mountains, increase soil erosion. Thin soil layer, which becomes loose, is transported from slopes by winter rains and, as a result of the process, naked limestone comes to the surface soon, making complete the erosion.

There are several examples for deforestation and cutting down trees in other regions, as well. In the reign of King Salamon, cedars made forests with a total territory of 5,000 km². Cedar forests were first mentioned in the literature between 2,500 B.C. – 2,300 B.C. However, recently very few cedars are found there. In the golden age of the Roman Empire the main road from Baghdad to Damascus was shadowed throughout by cedars. Recently, the road between these cities is surrounded by desert (*McNeill*, 2001).

Several cultures emphasize that one should live in harmony with the environment. However, even in those societies, where this idea has been perpetually mentioned (e.g. in Asian societies), environmental ideas got frequently lost on the surface of financial demands.

Air pollution problems of ancient times are mentioned even in poems of classical poets. Horatius (65 B.C. – 8 A.D.) wrote that Roman buildings became more and more dark from smoke and this phenomenon might have been observed in many ancient cities, as well. Seneca (4 B.C. – 65 A.D.), teacher of Emperor Nero (A.D. 37-68.), was in poor health all in his life and his physician frequently advised him to live Rome. In one of his letter, in 61 A.D, he wrote to Lucilius that he needed to live gloomy smoke and kitchen smells of Rome in order to feel himself better (*Heidorn*, 1978).

Roman law states that cheese making manufactures should be settled so that their smoke not to pollute other houses (*Mészáros*, 2002).

The Roman Senate introduced a law about 2,000 years ago, according to which: "Aerem corrumpere non licet", namely "Polluting air is not allowed."

LEAD MINING AND EXPLOITATION

In the ancient Mediterranean, mining and metallurgy played a basic role in economy. According to Xenophon (434-359 B.C.) and Lucretius (98-55 B.C.), harmful smoke of lead mines in Attica damaged health (*Weeber*, 1990).

Lead is extracted from its most important ore, namely galenite. Lead content of galenite is 86.6 %, and comprises yet arsenic, tin, antimony and silver. The most part of silver production of the world comes from galenite, and not from silver ore, since mining and exploitation of galenite is much more significant. A long time after introduction of silver coin as currency (about 7,000 B.C.), primary aim of mining galenite was to extract silver and lead was considered to be as a by-product (*Boutron*, 1995).

Lead mining started about 4,000 B.C. Considerable exploitation began one thousand years later, or so; when a new smelting technology was developed in order to extract lead (and silver) from sulphide ores of lead. Mass of exploited lead ore and use of lead became more and more important in the Copper-, Bronze- and Iron Ages (*Nriagu*, 1983a). This progress was promoted by the introduction of silver coins and development of Greek civilization (during that time lead production was 300 times higher than that of silver). Lead production reached its maximum of 80,000 tons/year in the golden age of the Roman Empire, which was about the same magnitude than that of the Industrial Revolution some 2,000 years later (*Hong et al.*, 1994). The most important lead mines were situated in the

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Iberian Peninsula, the Balkans, in the territory of the ancient Greece and in Asia Minor (*Nriagu*, 1983a). Lead production suddenly decreased after the fall of the Roman Empire and reached its minimum in the medieval ages with only a mass of some thousand tons/year. Then, the production began to increase again, due to the new lead and silver mines opened in Middle Europe since about 1,000 A.D.

USE AND APPLICATIONS OF LEAD

In the Roman times lead was the most popular metal and was widely used in the everyday life. Its compounds were used as face powders, lipstick or mask paint as well as colouring agent in paints. Furthermore, lead was used for preserving foods; even it was portioned to vine in order to prevent its fermentation. Lead compounds were used as a birth controlling medicine (for exterminating sperms) and a kind of a spice, too. Cups, jugs, pots and frying pans were made of lead alloys. Coins were also made of lead as well as of alloys of e.g. lead and other metals such as copper, silver and gold. Since it is resists corrosion and can be processed easily, lead was extensively used in shipbuilding, house building and water pipes were also made of it. During house building, hot lead was poured among limestone/marble blocks and, in this way, it served as binder. In the ancient Rome and in other cities of the Roman Empire, water pipes were the most important field of lead's application. Also, in Babylon a water pipe made of lead was used for watering the hanging garden built by king Nabu-kudurri-usur [with old style: Nabukodonosor (605-562 B.C.)]. Because of the above-mentioned facts, lead used to be mentioned as a Roman metal, too (*Markham*, 1994).

ILLNESSES CAUSED BY LEAD

Both lead and its compounds are poisonous. Little volatility (lead vapour) slight mouldering (lead powder) as well as volatility of some of its compounds [e.g. a petrol additive $(Pb(C_2H_5)_4)$] or solubility [e.g. $Pb(CH_3COO)_2$] make possible to get them into the constitution. Symptoms of lead poisoning are headache, nausea, diarrhoea, swoon and cramp.

Romans knew that lead is a dangerous metal, since they turned their attention to diseases of people working in lead mines. However, since it was used extensively in the everyday life, danger was taken out of consideration. Lead was believed to be less dangerous if it got into the constitution in little dose. Carbon dioxide molecules in water react with lead in the water pipes and the solution of lead compounds may enrich in the constitution; hence, they can possibly cause a so called "lead disease" a consequence of which might be paralysis. Lead in foods and drinking water might have led to infertility or still birth (*Goldstein*, 1988). Nevertheless, mineworkers suffered mostly from harmful effects of lead. Hence, Romans generally made slaves work in mines. In Greek-Roman times, according to an estimation, many hundred thousand people (mainly slaves) died in acute lead poisoning during mining and smelting processes (*Nriagu*, 1983a; 1983b; *Hong et al.*, 1994). It is possible that extreme manifestations of Emperors Caligula (12-41 A.D.) and Nero might also have been consequences of lead poisoning (*Goldstein*, 1988).

According to several researchers, one possible reason of the fall of the Roman Empire might have been the large scale occurrences of poisoning coming from the extensive lead mining and widely used devices made of lead (*Nriagu*, 1983a; 1983b; *Hong et al.*, 1994).

In the 20th century, lead compounds coming from $Pb(C_2H_5)_4$, which have been used for many decades as anti-knock additives, have caused pollution of the environment. Lead compounds, depositing from the atmosphere to agricultural plants, got into the constitution either through the food chain or when breathing. Petrochemistry could only recently change $Pb(C_2H_5)_4$ with another compound, which does not pollute the environment (*Boutron*, 1995).

LEAD POLLUTION OF ANCIENT TOOTH SAMPLES IN THE UNITED KINGDOM

English researchers, co-fellows of the Natural Environment Research Council and the British Geological Survey, studied the concentrations of lead in tooth enamel from Romano-British and early medieval people from various sites in the United Kingdom. Then, they compared the lead exposure of these people both with their prehistoric forebears and with modern people living in the United Kingdom today. A large study of the tooth enamel lead concentration of adults living in the United Kingdom, carried out in the early 1980s, found that contemporary people averaged about 3 parts per million (ppm). The great majority were also closely similar with little variation between the various localities studied. Some more recent analyses of modern children's teeth average around a few tenths of a ppm suggesting, as also indicated by the atmospheric data, that modern lead exposure may be falling. On the other hand Neolithic people, living before the use of metals, had tooth enamel lead concentrations that averaged 0.3 ppm. These concentrations are only a tenth of the average for modern people and possibly similar to modern children.

When analysing tooth enamel of Roman, Anglo-Saxon and Viking people living in the United Kingdom, researcher concluded that there were individuals with tooth lead concentrations greater than 10 ppm and even occasionally significantly more. Concentrations of this magnitude among modern people are associated with occupational or acute exposure and suggest that lead pollution was a significant problem for both Roman and early medieval ancestors of the British citizens.

The explanation may be the fact that England, Scotland, Wales and Ireland are all rich in natural lead deposits. Furthermore, each of these countries has abundant ores, which have been mined since antiquity. Probably, it was partly the riches of the country's lead ores – with their associated silver of course – which led to Rome's initial interest in the conquest of this most northerly reach of their Empire. It is familiar that the Romano-British, Anglo-Saxons and Viking people living in the United Kingdom were exposed predominantly to lead from ore sources, because of the characteristic isotopic composition of the lead remaining in their teeth.

On the other hand, high exposures were detected not only among people actively involved in lead mining, smelting or metal working, but in tooth enamel of children, too. This indicates that high lead concentration was considered an environmental rather than occupational problem.

LEAD POLLUTION ON REGIONAL AND HEMISPHERIC SCALES

In 1957-58, during the International Geophysical Year firstly started an extensive research programme to analyse information stored in many hundred thousand years old snow and ice layers of Greenland and the Antarctic. The aim of this research was to establish possible hemispheric scale air pollution for many thousand years old time periods. Later, the ice cores coming from this area served substantial information on the atmospheric effects of human activities (e.g. *Boutron et al.*, 1991, 1993, 1994). In Greenland, the deepest boring meets a period of 7,760 years, which is well before the age, when silver was first smelted from galenite. We can speak about background levels of the atmospheric lead concentration till this period (*Boutron*, 1995).

Chemical analysis of an ice core with 9,000 feet deep from Greenland (1 foot = 30.48 cm) made it possible to collect information on the atmospheric pollution of past ages back to 7,760 years. According to this, lead concentration in the atmosphere before beginning of lead production, when atmospheric lead came only from natural sources, was low. At this time the enrichment factor of the atmospheric lead was near 1 (0.8), which meant that this lead came from soils and rocks. 3,000 years ago lead concentration of the atmosphere practically agreed with levels measured at the beginning of lead production. This means that anthropogenic lead emission had yet been negligible till this time, considering the amount of lead coming to the atmosphere naturally. Atmospheric concentration of lead started to increase in the 5th century, and during the Greek-Roman times (between 400 B.C. and 300 A.D.) the enrichment factor of lead reached 4 and has remained at the same high level for 7 centuries. Namely, four times higher lead concentration was detected for this period in the snow and ice layers of Greenland comparing to the earlier, natural values. This has been the earliest detected hemispheric scale air pollution, almost 2,000 years before the industrial revolution and well before any other polluting effect (Hong et al., 1994).

In the golden age of the Roman Empire, about 2,000 years before, 5 % of the 80,000 tons total processed lead production got into the atmosphere, which might have resulted in an atmospheric emission peak of 4,000 tons/year (*Hong et al.*, 1994). Lead emission coming from metal processing caused an important local and regional air pollution all over Europe, which can be detected e.g. in lake deposits of southern Sweden (*Renberg et al.*, 1994). Furthermore, these emissions significantly polluted the middle troposphere over the Arctic (*Hong et al.*, 1994).

Rosman examined, where from lead pollution came in the ancient atmosphere. According to the analysis of lead isotopes ratios in ice cores, mines in the territory of Spain proved to be the main sources of atmospheric lead. These mines were supervised by Carthago between 535-205 B.C. and then they were followed by Romans till 410 A.D. About 70 % of lead in the ice layers of Greenland in the period between 150 B.C. –50 A.D. comes from the mines of Rio Tinto, in the south-eastern part of Spain (*Rosman et al.*, 1993).

During the Greek-Roman age, an important part of the fourfold increase of lead concentration in the middle troposphere over Greenland came from lead/silver mining and processing. During the Roman Empire, 40 % of the lead production in the world was resulted from Spain, Middle-Europe, Britain, Greece and Minor Asia (*Nriagu*, 1983a). Lead was smelted in open furnaces, at which the rate of emission was not checked. The leaving small aerosol particles, on the routes which have been well-known recently, could easily reach the Arctic region (*Hong et al.*, 1994).

After the fall of the Roman Empire, the atmospheric lead concentration suddenly dropped to the background level, which was characteristic 7,760 years ago. Then, in the Medieval and Renaissance Ages it began to increase again and 471 years before it reached double concentration than that detected during the Roman Empire (Boutron, 1995). Afterwards, the increase was continuous following the industrial revolution, too. From the 1930s till about 1960, snow and ice samples in Greenland indicated quick increase. This can be traced back to anti-knock additives of leaded fuels, which were used first in 1923 (Nriagu, 1990). On global scale, 2/3 of leaded additives were used by the Unites States in the 1970s, 70 % of which got directly into the atmosphere with exhaust gases of vehicles. Atmospheric lead concentrations measured in the 1960s were about 200 times higher than the natural values. This is one of the most serious, ever recorded, global scale pollutions of the environment on the Earth (Boutron, 1995). The sudden decrease, observed since 1970, can be traced back to increasing use of unleaded fuels. Recently, all petrol sold in the United States and its more and more increasing ratio in Europe is unleaded (*Nriagu*, 1990). Recently, Eurasia is responsible for 75 % of the atmospheric lead concentration on the Earth (Rosman et al., 1993).

Lead pollution of the atmosphere has been detected over the Antarctic since the beginning of the 20th century. Use of leaded fuels and then their forcing back can also be detected. Furthermore, it can be established that an important part of anthropogenic lead comes from South America (*Boutron*, 1995). At the same time, natural concentration changes of lead (and other heavy metals) were also considerable over the Antarctic during the past ages. Low concentration values were detected in the Holocene period, while lead concentration was two orders of magnitude higher than this during the last glacial maximum, about 20,000 years ago (*Boutron and Patterson*, 1986).

COPPER MINING AND EXPLOITATION

At the beginning (about 7,000 years ago), copper was produced from native copper. This has been the main procedure for about 2,000 years. Following this period, developing of smelting technique of oxide and carbonate ores as well as appearance of tin-bronze brought the real Bronze Age. Then, the production increased continuously. In the period between 2,700-4,000 before present, the total production was about 500,000 tons (*Tylecote*, 1976).

Copper production suddenly increased in the Roman times. In this period, copper alloys were used more and more degree both for military and civil aims (e.g. minting). Production reached its maximum 2,000 years ago with a mass of about 15,000 tons/year. In this period, the main copper mines were situated in the territory of Spain (half of the total production of the world resulted from this country, from the regions of Huelva and Rio Tinto), in Cyprus and Middle Europe (*Hong et al.*, 1996b). Total production, in the period between 2,250-1,650 before present, was about 5 million tons (*Healy*, 1988).

Generally, speaking about any metals, peak/decrease of the production goes together with the golden age / decline of the country. This establishment is valid for both the Roman Empire and China, as well. Decrease of mining of all metal ores, including copper, started with weakening of strength of the Roman Empire. After fall of the empire, copper production decreased significantly in Europe. World production has stagnated with a mass of about 2,000 tons/year until the 8th century and then started to increase again. This

increase, from European side, is especially due to opening of new mines in the 9th century in the territory of Germany and in the 13th century in Sweden (the latter particularly in the region of Falun) (*Pounds*, 1990).

Outside the Roman Empire, important copper production occurred in Southwest Asia and in Far East, too. When the Han dynasty (206 B.C. –220 A.D.) extended its influence to Southwest Asia, copper production of China was about 800 tons/year. In the medieval age, most part of the world production came from China (during the rule of the northern Sung dynasty). In this period, the Chinese production reached its maximum of 13,000 tons/year and this resulted in the peak of the world production of 15,000 tons/year in 1080s A.D. Most part of copper was used for minting (*Archaometallurgy Group, Beijing University of Iron and Steel Technology*, 1978). During some hundred centuries after this period, the production suddenly dropped (about 2,000 tons/year in the 14th century) and then started to increase again from the industrial revolution till recently. (A comparison: the total copper production of the world was 10,000 tons/year at the beginning of the industrial revolution.)

COPPER POLLUTION ON REGIONAL AND HEMISPHERIC SCALES

Before the beginnings of anthropogenic use of copper, about 7,000 years ago, the total atmospheric copper came from natural sources and the situation has not changed even until 2,500 years ago. Beginning from 2,500 years ago, the atmospheric copper concentration has increased, which is a consequence of large scale copper pollution in the northern hemisphere (*Hong et al.*, 1996a).

Copper emissions from the ancient times to the recent period have been resulted from mining and metallurgical activities. Other anthropogenic activities (e.g. production of iron and non-ferrous metals, wood burning) contribute to these emissions only to a lesser extent.

Emissions concerning the production, in connection with a significant technological development, have considerably changed during the past 7,000 years. In the ancient times, due to the primitive smelting procedures, the emission factor was about 15 %. At the beginning, several steps of processing of sulphide ores (roasting, smelting, oxidation, cleaning) were performed in open furnaces. Emission has been taken out of consideration until the industrial revolution. From this time on, more developed furnaces and more recent metallurgical procedures have spread. Since the middle of the 19th century, the processing procedure has reduced to five steps. These technological developments have resulted in significant decrease of the emission factor. In the 20th century, this factor has only been 1 % and later, with introducing further modifications, it became a mere 0.25 % (Hong et al., 1996a; 1996b).

Since the Roman times the Cu/Al ratio has increased in ice samples, which indicates that in this period considerable copper pollution occurred in the troposphere over the Arctic. This copper might have originated during the high temperature section of the processing as small-sized aerosol particles and got into the atmosphere. These aerosols can easily reach the Arctic region leaving middle latitudes, where from they are resulted (in the Roman times: mainly the Mediterranean Basin, especially Spain; in the medieval ages: China).

Change of the Cu/Al ratio in ice samples meets the estimated change of anthropogenic copper emission. Data resulted from ice cores in Greenland indicate low values until 2,500 years before, medium values from the Roman times until the industrial revolution and suddenly increasing values near the recent period. Data from the Roman times show high variability. This can probably be traced back to the fact that in this period production of copper occurred in short periods, in the function of how many copper coins were needed (*Hong et al.*, 1996a).

According to the ice samples in Greenland, comparing production data with emission factors, atmospheric copper emission culminated twice in the period before the industrial revolution. The first peak occurred in the golden age of the Roman Empire about 2,000 years ago with a mass of some 2,300 tons/year, when use of metal coins spread in the Ancient Mediterranean. The second peak appeared in the golden age of the northern Sung dynasty (960-1279 A.D.) in China, at about 1080 A.D. with a mass of some 2,100 tons/year, when the Chinese economy was extensively developing and copper production increased. Since smelting technology was primitive at that time, about 15 % of the smelted copper got into the atmosphere. Though the total copper emission of the Roman and Sung times was about a tenth of that in the 1990s, copper production did not reach even a hundredth of that in the recent period. Hemispheric copper pollution caused by copper emissions has more than 2,500 years old history and copper emissions of the Roman and Sung times were so high than never before 1750 (*Hong et al.*, 1996b).

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