# Comparative review of croatian and indian air pollution studies with emphasis on pollutants derived by coal combustion

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# Comparative review of Croatian and Indian air pollution studies with emphasis on pollutants derived by coal combustion

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Review scientific paper

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## Abstract

Human health, the environment, and climate are being profoundly affected by complex pollutant interactions in the atmosphere. Almost all human activities cause the emissions of air pollutants. Their understanding and quantification is the first step towards the control and mitigation of air pollution. The general aim of this paper is to summarise the findings of selected Croatian and Indian papers addressing air pollution, particularly those focusing on sulphur and aerosols associated with coal-fired power plants. The two countries are essentially different regarding their size, geography, history, economy, industrial potential, to name but a few. However, they both have had certain relations to the fossil fuel extraction and its usage in power stations and industry for decades. Various research approaches are presented, together with a brief outline of national air pollution policies.

## Keywords

sulphur, aerosols, coal combustion, air pollution, atmospheric research

## 1. Introduction

Croatia is a country located in Southeastern Europe, having a total area of 56,542 km$^2$. The length of its boundaries with six neighbouring countries (Slovenia, Hungary, Serbia, Bosnia and Herzegovina, Montenegro, and Italy on the Adriatic Sea) is 2,375 km. Its climate in the lowlands is characterised by hot, dry summers and cold winters, while in the mountains, summers are cool and winters are cold and snowy. Along the Adriatic coast, the climate is Mediterranean with mild winters and dry summers. The main environmmental problems include air pollution, deforestation, and contamination of coastal waters with industrial and domestic waste. Due to the decentralisation of environmental management, cities and municipal administrations determine environmental policy. The Croatian major air pollution source is situated in the Labin city area which belongs to the Istrian Peninsula (Northern Adriatic Coast, see **Figure 1**). The largest Croatian coal-mining area is located there, and was excavated as early as the 17th century, while its termination was occurring during the late 1980s. The sole Croatian coal-fired power plant Plomin is also situated there, which started with operation in 1970 using the domestic Raša coal which contained up to 13% of sulphur (S) (Medunić et al., 2016a, b), while imported low-S coal has been used for the last 15 years. Croatia's electric power generating capacity totals 10,500–14,500 GWh, of which nearly two thirds is hydroelectric and one third is from conventional thermal sources. To meet its need for coal, 1.1 million tons of coal was imported in 2010. Since Croatia is one of the European Environment Agency's (EEA) member countries, its air pollutant emissions and projections are published in the form of a fact sheet (EEA, 2014), which presents compiled information based on the latest official air pollution data reported by the EES member countries. This document has reported generally decreasing $SO_2$ emissions (generated from energy use, 93%) since 1990 (170 Gg $SO_2$; 1 gigagram (Gg) = $10^9$ g = 1 kilotonne (kt)) till today (20–30 Gg $SO_2$), thus complying with the National Emission Ceilings Directive (NECD) emission limit for the year 2010 set at 70 Gg. Regarding fine particulate matter ($PM_{2.5}$), originating mainly from energy use (68%), road transport (17%), and industrial processes (11%), the general trend during the 1990–2012 period has been -3 Gg (-24%), whilst distance of the latest year $PM_{2.5}$ emission data to emission ceiling in 2020 is 1 Gg (9%). The behaviour and fate of air pollutants in the environment, together with the human exposure to air pollutants in Croatia are investigated and monitored by the Institute for medical research and occupational health (IMI). Their scientists regularly publish results in scientific journals and conference proceedings (e.g. Fugaš et al., 1999; Vađić,

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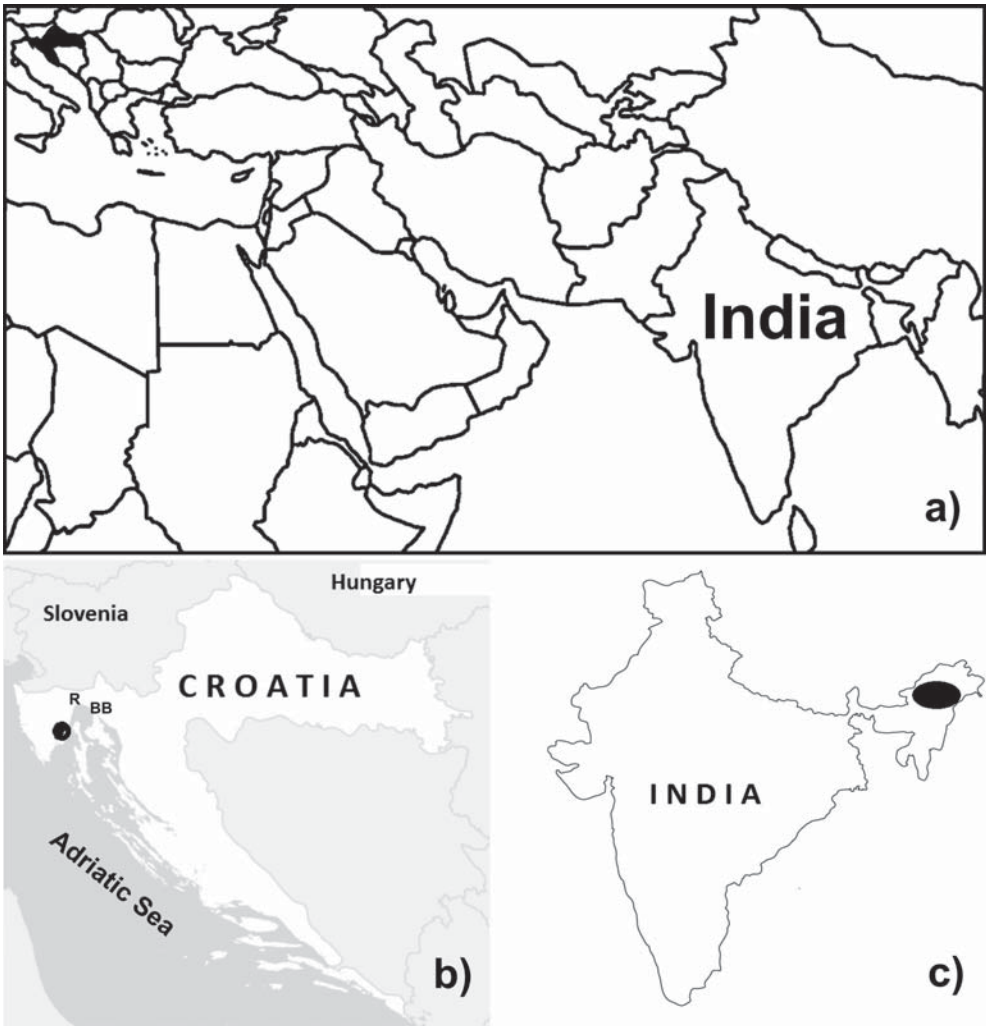

**Figure 1.** Geographic position of Croatia and India. a) Schematic presentation of Croatia (black colour, left upper part of the graphic), and India; b) Black dot - the position of the Labin city and the Plomin coal-fired power plant; R – the city of Rijeka; BB - the Bakar bay; c) Black dot - the position of the Assam coal deposits.

2003). Regarding the coal-fired power plant Plomin, although the sulphur emissions have dropped significantly with the new block B since 2000, Božičević Vrhovčak et al. (2005) noted that the highest annual $SO_2$ emission still originates from the block B (7015 tons, or 5007 mg/kWh). Furthermore, the authors investigated damages to human health resulting from the Croatian thermal power plants' annual operation. Following the use of GIS to combine the data on the population density and calculated ambient concentrations of PM, they found out that the block B, having occupied the second place on the list of power plants' shares, accounted for 27% of the total impact. The most notable health endopoints were associated with chronic cough and chronic bronchitis for children, and chronic mortality. Božičević Vrhovčak et al. (2005) also determined that the second largest annual (year 2000) total costs (6,480,000 USD) of human health degradation were caused by the Plomin block B.

The Republic of India is Asia's second-largest country after China, filling the major part of the South Asian subcontinent together with Pakistan, Nepal, Bhutan, and Bangladesh. Also, its territory includes the Andaman and Nicobar Islands in the Bay of Bengal, and Lakshadweep (formerly the Laccadive, Minicoy, and Amindivi Islands) in the Arabian Sea. The total area is 3,287,590 km$^2$, i.e. nearly 60 times larger than Croatian territory. India is bordered by following countries: China, Nepal, Bhutan, Myanmar, Bangladesh, and Pakistan. The total boundary length is 21,103 km, i.e. nearly 9 times Croatian boundaries, of which 7,000 km is coastline. Unlike Croatia's rather simple climate pattern, the climate of India comprises a wide range of weather conditions across a vast geographic scale and varied topography, making generalizations difficult. India hosts six major climatic subtypes, ranging from arid desert in the west, alpine tundra and glaciers in the north, and humid tropical regions supporting rainforests in the southwest and the island territories, while many regions have starkly different microclimates. Its main environmental problems are land damage, water shortages, and air and water pollution. Because of the rapid industrialization and urbanization in recent years, air pollution has acquired critical dimensions and the air quality in most Indian cities that monitor outdoor air pollution fail to meet the WHO (World Health Organization) guidelines for safe levels. The levels of $PM_{2.5}$ and $PM_{10}$ as well as concentration of dangerous carcinogenic substances such as sulphur dioxide and nitrogen dioxide have reached alarming proportions in most Indian cities. Air pollution is most severe in urban centers. Two decades ago, India had the world's sixth-highest level of industrial carbon dioxide emissions, which totaled 769 million metric tons. According to a study conducted by Greenpeace India in 2011−2012, emissions from Indian coal-fired plants resulted in 80,000 to 115,000 premature deaths and more than 20 million asthma cases from exposure to total $PM_{10}$ pollution. According to the latest urban air quality database released by the WHO, India ranks among the world's worst for its polluted air. Out of the 20 most polluted cities in the world, 13 are in India. The report reconfirms that most Indian cities are soon becoming death traps because of very high air pollution levels (Times of India, May 9, 2014).

## 2. General facts about coal

Coal is combustible sedimentary rock formed from vegetation that has been consolidated between rock strata for millions of years (Speight, 2005). The initial reactions in the coalification process involve the microbial degradation of plant residues, either aerobically or anaerobically, into humins and peat. Increased pressure and temperature alter the physical and chemical characteristics of the resulting sediment, which is transformed into coal. Due to the heterogeneity of plant tissue and varying geochemical conditions, the structure of coal will differ between coal seams (Kirby et al., 2010). Almost all the elements of the Chemical Periodic Table can be found in coal. According to their different contents, these elements can be divided into three groups: (1) major elements (C, H, O, N, S), the amounts of which are above 1000 mg/kg; (2) minor elements, those associated with mineral matter (Si, Al, Ca, Mg, K, Na, Fe, Mn, Ti), and halogens (F, Cl, Br, I), present in concentrations between 100 and 1000 mg/kg; and (3) trace elements, e.g. Cu, Zn, Pb, Cr, Ni, Hg, Se, As, etc., contributing with concentrations below 100 mg/kg (Swaine and Goodarzi, 1995; Xu et al., 2003; Rađenović, 2006). Trace elements were concentrated by processes which took place prior, during, and following the formation of coal. A variety of factors influence the trace element content of coal, e.g.: concentration of trace elements during growth of vegetation; enrichment of trace elements during the decay of plant material; sedimentation and diagenesis; burial and coalification; and subsequent mineralization (Clarke and Sloss, 1992). The trace elements may be associated with certain groups that are part of the organic matter, as are carboxylic (-COOH), phenolic hydroxyl (-OH), mercapto (-SH), and imino (=NH). Likewise, trace elements are mainly associated with mineral matter, as discrete minerals, either free or embedded in the organic matter, as replacement ions in minerals, and adsorbed on minerals (Finkelman, 1994; Swaine and Goodarzi, 1995; Vejahati et al., 2010). These elements mostly evaporate during combustion, and condense either homogeneously as sub-micron ash or heterogeneously onto already existing fine ash, the former one being more difficult to be captured. Clean coal technology development is, therefore, a priority area for research and needs continuous improvements in increased efficiency and decreased pollutant emission (Swaine, 1994; Swaine and Goodarzi, 1995).

In most cases, oal ashes have high concentrations of trace elements when compared to other geological materials (Ketris and Yudovich, 2009). During combustion or gasification, coal particles undergo complex changes, including the formation of char, agglomeration of melted inclusions and vaporization of volatile elements (Clarke, 1993). Trace elements are important because of their association with environmental issues and the health of plants, animals and humans (Finkelman, 2004). Consideration must be given to essentiality, non-essentiality, and toxicity; they all depend on concentrations, element forms (speciation), pH and oxidation–reduction condition. In some cases, the difference in concentration between essentiality and unwanted effects, even toxicity, is small (Swaine, 2000). The major components in coal ashes are silica (20–60%$SiO_2$), alumina (5–35% $Al_2O_3$), ferric oxide (10–40% $Fe_2O_3$), and calcium oxide (2–10% CaO) (Flues et al., 2013). Throughout the coal combustion, ashes enriched 3–10 times in trace elements are produced and emitted into the environment.

## 3. Sulphur in coal - abundance and forms

The sulphur content in coal varies, most commonly ranging from 0.5% to 5% total sulphur. Coals with less than 1% sulphur are classified as low-sulphur, 1% to 3% sulphur are medium-sulphur, and ≥3% sulphur are high-sulphur coals (Chou, 2012). Sulphur is the most abundant heteroatom in coal. The sulphur content and forms vary considerably with location and coal rank. Sulphur in coal appears in both organic (sulphur bonded to the hydrocarbon matrix), and in inorganic (metal sulphides, disulphides and sulphates) forms, as well as elemental sulphur (Kasrai et al., 1996). Elemental sulphur is not present in pristine coal, but primarily derives from the oxidation of pyrite. It is generally present in relatively small amounts even in oxidised coals. The iron disulphide, $FeS_2$, appears in two crystalline forms, the pyrite (cubic) and the marcasite (rhombic). Pyrite is the predominant sulphide mineral in coal. Other sulphide minerals found include marcasite, pyrrhotite, sphalerite, galena, and chalcopyrite, as well as rare occurrences of getchellite and alabandite. The sulphate sulphur appears mainly in the form of calcium and iron sulphates. The organic sulphur compounds in coals may be classified into three categories: 1) thiols, 2) sulphides and disulphides, and 3) thiophene and its derivatives. The sulphur-containing aromatic compounds (benzothiophene, dibenzothiophene, and benzonaphthothiophene) were found in bituminous coal and anthracite, but not in lignite. Thus, the abundance of various types of organic sulphur compounds in coal may be related to the rank of coal (Rađenović, 2004; Chou, 2012). The superhigh-organic-sulphur coals are highly enriched in organic sulphur, usually in the range between 4% and 11% (e.g. Raša coal, Croatia). High-sulphur coal is enriched in certain trace elements relative to low-sulphur coal (Kolker, 2012). Variation in sulphur content of coal is controlled mainly by geological conditions. Much of the sulphur in low sulphur coal derives from the sulphur content of the plant material making up the original peat. Sulphur contents greater than a few tenths of a percent have long been known to derive from the depositional environment. Sea water or brackish water in the coal beds contains sulphates. The sulphates undergo bacterial reduction to $H_2S$ which reacts with iron in the water to form pyrite, and with the organic material or the sulphate reducing bacteria to form the organic sulphur structures (Dai et al., 2002). The range of organic sulphur contents in exinite is much wider than that of other macerals (Guijian et al., 2001). In contrast to inorganic sulphur (such as pyrite), which can be isolated by physical methods, organic sulphur, being part of the coal structure, cannot be removed by physical methods. Organic sulphur in coal is traditionally calculated as the difference between the total sulphur and the sum of pyritic plus sulphate sulphur (Attar, 1978).

## 4. Air pollution associated with coal combustion processes

The fossil energy sources include coal, petroleum, bitumens, natural gas, oil shales, and tar sands. These fuels have been formed in the geological past and are not renewable. The major non-fuel use of coal is carbonisation for the purpose of making the metallurgical coke. The production of activated carbon from coal has been of interest for many years. Carbon in coal can be used as a source of special aromatic and aliphatic chemicals via processing, including gasification, liquefaction, direct conversion, and coproduction of chemicals, fuels and electricity (Ghosh and Prelas, 2011). In the future, coal will keep its important position as a world energy source because of its relatively abundant reserves in comparison to the decreasing reserves of both petroleum and natural gas (Vejahati et al., 2010).

The pollutant emissions from coal utilisation may cause serious environmental and health problems (Nriagu, 1990; Xu et al., 2003). The emissions of $CO_2$, SOx, NOx, and some volatile inorganic elements (As, Be, Cd, Co, Cr, Hg, Mn, Ni, Pb, Sb, Se), and their compounds in flue gases from coal combustion, may have important environmental impacts such as global temperature rising, and direct hazards of volatile compounds to agriculture, soil, water, and human health. Amongst them, sulphur is the most notorious environmental contaminant, resulting in acid rain (Kolker and Finkelman, 1998; Sparks, 2003; Rađenović, 2006; Kampa and Castanas, 2008), and also acid-mine drainage problems (Burgos et al., 2012). Numerous studies have shown that coal-fired power plants are one of the largest global sources of pollution that pose a potential threat to the environment and human health (Laumbach et al., 2015). Various inorganic as well as organic contaminants are released by the coal combustion. The most important contaminants are sulphur and nitrogen oxides, carbon dioxide, fine-grained particulate matter ($PM_{2.5}$ and $PM_{10}$), potentially toxic heavy metals (La, Ce, Hg, Te, Th, Cr, Hf, Sc, Zn, Fe, Tl, Co, Sm, Am, As, Se, Be, Cd, Pb, and Mn), PAHs, and other combustion products (Clarke and Sloss, 1992; Querol et al., 1995; Llorens et al., 2001). In addition to endangering ecosystems at local and regional levels, coal-fired power plants emit large amounts of carbon dioxide, which is responsible for the global problem of climate change (Duić et al., 2005). The adverse effects on health and the environment due to the emission of gases and particles from coal-fired power plants can vary in time and space due to various factors, such as local geology, demography, climate, etc., so the negative effects of power plants will vary from place to place. The important geological features that determine the damage caused by plants are the permeability and thickness of the soil, the type of bedrock, the mineral composition of rocks and soil, and relief. At a local level, the greatest impact is related to deposited ash, while gases have regional, and even global negative impact. Fly and bottom ash remain after coal combustion, commonly containing heavy metal levels elevated compared to the original coal. If the ash is not appropriately stored, its heavy metal load can reach the ground water, and through the plants enter the food chain (Fernández-Turiel et al., 1994; Oreščanin et al., 2009a, b; Khillare et al., 2012). Among the gases, sulphur in particular has been associated with the principal environmental problems confronting society which is heavily dependent on coal derived electricity. Aside from its role in the acidification issue and concomitant environmental pollution (Gorham, 1976), sulphur is a critical nutrient (Zgorelec et al., 2012) as well as an important participant in regulating climate (Drake, 2011). Hereby, the knowledge of the fate of sulphur in the ecosphere should be a matter of the utmost importance.

The vast amounts of studies have been devoted to emissions and behaviour of trace elements and sulphur in coal (e.g. Mukherjee and Srivastava, 2005; Baruah and Khare, 2010; Verma et al., 2015), whilst beneficial or detrimental ecological effects of sulphur (Oden, 1976; Puklińska et al., 2013), its reservoirs in nature, as well as challenges of clean-up campaigns (Álvarez-Ayuso et al., 2006; Dowarah et al., 2009) are fairly lacking.

## 5. Croatian air-pollution studies

The air quality monitoring in urban and industrial areas in Croatia started in Zagreb during the 1960s, carried out by the Institute for Medical Research and Occupational Health and regional authority (Vađić, 2003). Since the early 1970s, air pollution monitoring had been gradually introduced in other Croatian towns by regional Institutes of Public Health together with the regional authorities. All regional monitoring networks use the same air quality monitoring methodology, being connected in one common network which is the sole monitoring network in Croatia. Some local monitoring stations are specifically located in industrial areas, gas fields, thermal power plants, and near the waste dumps. Otherwise, global indicators of air quality in Croatia are monitored by Meteorological and Hydrological Service. An ongoing air quality surveillance in the regional network is focused on levels of sulphur dioxide, smoke, total suspended particulate matter (SPM), lead (Pb), cadmium (Cd), and manganese (Mn) in total SPM, PM10, nitrogen dioxide, ozone, and PAH measurements in the seven largest Croatian towns. According to Vađić (2003), the results indicate that the air in Croatian towns has been moderately polluted with SPM, PM10, NO2 and BaP (benzo(a)pyrene). Levels of Pb, Cd, and Mn in SPM, and ozone (O3) have been very low during the whole surveillance period. Generally, Croatian towns follow trends similar to those of other European countries. For more information about relevant research studies readers are suggested to consult following references (Čačković et al., 2008, 2009; Šišović et al., 2008, 2012; Žužul et al., 2011; Zgorelec et al., 2012; Jakovljević et al., 2015).

Papers addressing air pollution around the cities of Rijeka (great industrial centre), and Plomin (a sole Croatian coal-fired power plant) are briefly presented in the following text. Šinik et al. (1994) carried out a study measuring a simultaneous hourly series of $SO_2$ concentration and wind velocity in the Bakar Bay on the Northern Adriatic coast (Fig. 1b). They defined the local background pollution as the minimum pollutant concentration which cannot be cleared away when wind velocity and turbulent diffusivity approach their maximum. It was found that the greatest background $SO_2$ concentration was connected with winds blowing from ESE-SSE directions, with the air streams along the Croatian coast of the Adriatic Sea. Matković and Alebić-Juretić (1998) presented results of air monitoring in the city of Rijeka (Fig. 1b), which was one of the most polluted cities in Croatia as regards to $SO_2$ due to its high emission from industrial plants. Annual means of $SO_2$ exceeded 100 μg/m$^3$ in the city centre, 70−80 μg/m$^3$ in the Bakar Bay area (Fig. 1b), and 40 μg/m$^3$ in the suburban residential area, whilst the guideline value was 50 μg/m$^3$. The paper shows trends in $SO_2$ annual mean concentrations in the period 1986−1995 for two urban, two industrial, and one suburban site. Compared to 1989, there was a reduction of nearly 71% of $SO_2$ emission in 1995. The reasons were partial use of gas for energy supply in a petroleum refinery and some municipal heating plants, low sulphur fuel, and a reduced production in some industrial plants. Regarding the dustfall measurements in the wider area of the city of Rijeka, they started in 1975 in Bakar Bay, followed by the city of Rijeka in 1982, the nearby islands Krk and Cres in 1986, and Gorski Kotar in 1995 (Mićović et al., 2010). Generally, the recommended and limit values were only occasionally exceeded in Bakar Bay due to emissions from the coke plant, and the harbour as well as from a local shipyard. Moreover, deposition of S and N at the inland sites within the mountainous area, repeatedly claimed as influenced by acidic deposition, was below the respective critical load values, thus not responsible for the observed forest decline. Since the Bay of Bakar (Fig. 1b) has been one of the most heavily polluted bays of the Eastern Adriatic, endangered by three major industrial companies, Popadić et al. (2013) discussed major, minor, and trace element levels in surface sediments in relation to the sediment type and foraminiferal assemblages. They found that the area in front of the coke plant and the city of Bakar harbour was heavily polluted with some elements, chiefly with As, and Ni. Furthermore, they determined that stress-tolerant foraminiferal species dominated stations which were characterised by increased heavy metal levels. The Plomin city area (Fig. 1b) is an interesting site from an environmental point of view as it has been a major source of the Croatian energy production for more than 100 years (Medunić et al., 2016b). As the domestic Raša coal was characterised by remarkably high values of sulphur, up to 13−14% (Hamrla, 1959; Valković et al., 1984a, b), a few medical studies (carried out by Croatian gynaecologist L. Mohorović) established the correlation between ground $SO_2$ levels and health problems of pregnant women and small children (Mohorović, 2003, 2004). Potočić et al. (2003) conducted the 15-year monitoring of $SO_2$ emitted by the Plomin power plant with the black pine needles at different distances from the plant. The results showed that their S concentrations decreased approximately two years following a significant drop in emitted $SO_2$ levels. Regarding soil around the plant, it was found to be severely contaminated with S and PAHs (Medunić et al., 2014, 2016a) as well as with Ra-226 (Ernečić et al., 2014), while peculiar REE patterns were found (Fiket et al., 2016). PAHs are typical products of anthropogenic processes (De Nicola et al., 2015). Their total concentrations in topsoils varied from 31 ng/g in the control samples, to 13,535 ng/g at a distance of 100 m from the power plant (Medunić et al., 2016a). The authors have concluded that the major sources of PAHs in the Plomin area have been the coal-combustion processes, whilst the additional contribution could have been from a nearby unburnt coal pile subjected to atmospheric-dispersion processes.

Since the Raša coal had the increased radioactivity (Hamrla, 1959), the study carried out by Bauman and Horvat (1981) investigated working individuals exposed to enhanced levels of naturally occurring radionuclides in the Plomin power plant, since their surface contamination was inevitable as the radioactive dust penetrated everywhere. The authors determined increased Pb levels in urine and chromosome aberrations. They concluded that the hazards from exposure even to low doses of natural radiation by intake, inhalation, and surface contamination could not be neglected. Also, Lokobauer et al. (1997) presented the results of a preliminary investigation of radon activity concentration in houses around the plant Plomin. It was conducted in the winter of 1990 and the spring of 1991, when the difference between radon levels in old and new houses was noted for the first time, which was followed by the subsequent radon measurement data in selected houses during the period 1992–1994. The assumption was that some old houses were built using mortar and plaster that contained ash and slag from the coal combustion. The authors estimated that the average annual effective doses from inhalation of radon progeny for the inhabitants living in old and new houses were 2.7 mSv and 0.7 mSv, respectively. The study (Marović et al., 2004) assessed the radiological situation in the area of the Plomin power plant related to a waste landfill located near the sea and a very populated region. The waste is the coal combustion residue, i.e. TENORM (technologically enhanced naturally occurring radioactive material), characterised by increased radioactivity due to elevated levels of natural radionuclides in domestic coal. Systematic radioactive measurements prior to and following the remedial activities showed that the ash and slag waste was well monitored, and that the calculated absorbed dose rate significantly dropped in the latter case. Moreover, papers addressing damage to human health resulting from the annual operation of eight Croatian thermal power plants were also published (Božičević Vrhovčak et al., 2005; Strijov et al., 2011). In 1997 the Croatian government has prescribed the limit values on airborne emissions stemming from stationary sources (Official Gazette, 1997), as presented in Table 1. According to Božičević Vrhovčak et al. (2005), the majority of Croatian thermal power plants did not satisfy the requirements of the by-law ten years ago.

**Table 1.** Limit values of pollutant emissions from stationary sources over 500 MW from thermal power plants in Croatia, in mg/m$^3$ (Official Gazzete, 1997)

| | $SO_2$ | $NO_2$ | PM |
|---|---|---|---|
| **Coal** | 400 | 650 | 50 |
| **Oil** | 400 | 450 | 50 |
| **Gas** | 5 | 350 | 5 |

## 6. Croatian air-quality legislation

Nećak and Barbalić (2009) extensively elaborated air-quality legislation in Croatia. Basically, policy and measures for air quality protection and improvement in the Republic of Croatia are regulated by the Environmental Protection Act (OG 80/13), and Air Protection Act (OG 130/11). The latter prescribes air quality assessment, air quality monitoring, emission monitoring, emission limit values of emissions from stationary sources, limit and critical levels of pollutants in the air, requirements on technical facilities and fuel, and inspection and quality of measurement data. The Ministry of Environmental and Nature Protection (MENP) is the central governmental body responsible for the implementation and monitoring of the protection and improvement of air quality at the national level. It is the competent authority regarding the cooperation with other member countries and the European Commission. For the purpose of efficient management of air quality, MENP in cooperation with relevant central state administration made plans for the protection and improvement of air quality and action plans, national programs and national reports. Hereby, these actions have been made in order to meet obligations assumed under international treaties in the field of air and the competent authority for monitoring the implementation of these documents in the Republic of Croatia. The documents are issued by the Government of the Republic of Croatia.

## 7. Indian air-pollution studies

Amongst the coal producing countries, India ranks third. Its production of coal equalled to 431.27 million tonnes in 2009–2010 (Patra et al., 2012). Coals from the Northeastern part of India (Fig. 1c) have been characterised by high sulphur content, 2.5–6%, which makes them unsuitable for metallurgical or domestic applications. Their use would cause rapid corrosion of metal parts and contribute to the environmental pollution. Also, they have low ash, and high volatile matter. Therefore, these coals have attained a considerable attention of Indian scientists during the past 2-3 decades.

Mazumder et al. (1989) carried out desulphurisation of high-sulphur coals at normal atmospheric pressure and at elevated pressures. Using sodium compounds of both straight chain and aromatic alcohols, they achieved a reasonable rate of organic sulphur removal. Pyritic and sulphate sulphur were removed to the extent of 90% or more in almost all cases. The treatment time was also relatively small. The authors also discussed consequent changes in caking index, calorific value and softening point. The dependence of the extent of sulphur removal on particle size of coal was also studied. The authors noted that the expenses of the process were low as most of the spent solvent could have been regenerated. Baruah and Khare (2007) conducted pyrolysis experiments under laboratory conditions on five high sulphur coal samples from the states of Meghalaya and Nagaland (India), at temperatures of 450, 600, 850, and 1000°C. The yield of products and thermal release of sulphur from the coal samples were investigated. The distribution of sulphur in the pyrolyzed products, i.e., char/coke, gas, and tar, was also reported. Hydrocarbon and sulphurous gases released at different temperatures were analysed by gas chromatograph (GC) with an FID (flame ionized detector) and FPD (flame photometric detector), respectively. Maximum sulphur release was found in the range of 600–850°C, having a decreasing tendency from 850–1000°C, which might have been due to the incorporation of sulphur released into the coal matrix. Coals from the Northeastern region of India are characterised by high sulphur, low ash, and high volatile matter.

The study conducted by Khare and Baruah (2010a) was focused on sulphur, metals and ash contents of feed coals so as to make their emission inventory. For the calculation of emission factors of $SO_2$ and metals, mass balance method was used, while measured values near the coke ovens were used for $PM_{2.5}$. The emission factors and emission rates of $SO_2$ for coke ovens ranged between 0.80 to 4.8 Kg/t, and 204 to 1226 t/yr, respectively. The emission factors for $PM_{2.5}$, total carbon, black carbon and organic carbon varied between 0.7 to 3 Kg/t, 0.48 to 2.1 Kg/t, 0.007 to 0.03 Kg/t, and 0.47 to 2.1 Kg/t, respectively. The emission rates of metals (V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Mo, Cd, Sn, Te, Hg, Pb, and Bi) showed dependency on the volatility of the metals, condition of coke ovens (reductive) and rank of coal. The study has provided preliminary information on source profile of coke oven emissions ($SO_2$, $PM_{2.5}$ and trace metals) which is useful for an assessment of the impact of coke oven on ambient air quality, source apportionment, and the coke oven design so as to regulate future emissions. In the study by Khare and Baruah (2010b), aerosol $PM_{2.5}$ samples were collected at an Indian suburban site, exposed to different source emissions such as vehicular emissions, wood burning, coal based industries and other industrial activities, during the 2007–2008 period. The mass concentrations of $PM_{2.5}$, major elements (Al, Si, P, S, Na, K, Ca, Ti, V, Cr, Mn, Fe, Te, Co, Ni, Cu, Zn, Cd, Sn, Sb, and Pb), and major ions ($Cl^-$, $NO^{3-}$, $SO_4^{2-}$, and $NH^{4+}$) were determined for winter and rainy seasons. Their levels were found to be higher compared to various European and American cities, yet comparable to the Asian cities. The source identification of the study showed that $PM_{2.5}$ levels were influenced not only by local and industrial activities, but also by the long range transport.

Khare and Baruah (2010c) carried out chemometric analysis to evaluate the release behaviour of trace elements during coal utilization processes. They applied principal component analysis (PCA) and linear discriminant analysis (LDA) on the TE concentrations of raw and thermally treated coals. The PCA and LDA successfully predicted the association of 21 trace elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Sb, Te, Pb) contained in coal and their thermal behavior at various temperatures. By the application of chemometric on thermally treated coals it was possible to show that at 450°C, elements like Na, P, K, Fe, Ca, Mg, Al, and Si had an affinity for the mineral matter. Elements like Te, Sb, and Ti may form the chlorides which enhance their volatilities, while Co and Pb may form sulphides. In the temperature range of 600–850°C, coal had undergone an intense degradation of its structure during pyrolysis, whilst the elements released may have been adsorbed on coal surface or be volatile. The authors found out the results of chemometric analysis were in good agreement with volatilities of TEs present in coals at various temperatures, as well as with the FTIR analysis.

Khare et al. (2011) conducted research on $PM_{2.5}$ and $PM_{10}$ samples collected from a suburban site in the NE part of India; they analysed particle mass, total carbon, water-soluble total carbon, water-soluble organic carbon, water-soluble inorganic carbon, organic acids (formic, acetic, proponoic and oxalic acids) along with inorganic ions ($NO^{3-}$, $SO_4^{2-}$ and $NH^{4+}$). Based on absolute principal component analysis, four factors were resolved, associated with carbonaceous aerosols released from combustion of coal and wood, secondary inorganic and organic aerosols, and water-soluble inorganic fraction. Khare et al. (2012) investigated the characteristics of emissions ($PM_{2.5}$, $PM_{10}$ and suspended particulate matter), $NO_2$, $SO_2$, and $NH_3$ during coal utilization processes (carbonization and combustion) and mining. They compared emissions with those released from the combustion of wood along with the background concentration. Furthermore, they carried out morphological characterization, organic functionality, elemental, and ionic composition of particulate samples. The study showed that the gases released had depended upon the fuel utilization processes and their precursor concentrations in fuel. The size distribution and ionic composition of particles had depended upon the fuel types and processes of particulate formation, i.e., nucleation and agglomeration. Also, they determined that the element distribution in coal and their association controlled the emission of particulate bound elements.

Among the other Indian studies, some of them aimed at mineral characterisation of Assam coal (Saikia and Ninomiya, 2011), coal desulphurisation by the indigenous fungal culture isolated from coal (Acharya et al., 2005), soil contamination with heavy metals around coal fired power plants (Agrawal et al., 2010), distribution patterns of PAHs in fly ash (Sahu et al., 2009), vegetation (Sharma and Tripathi, 2009), and air (Saikia et al., 2016a), mineralogical and biological characterisation of fly ash (Saikia et al., 2015), and environmental and toxicological assessment of aerosols (Saikia et al., 2016b).

## 8. Indian air-quality legislation

To control the air pollution, the Indian parliament enacted The Air Prevention and Control of Pollution Act in 1981. Its objective was to provide the prevention, control, and abatement of air pollution. Decisions were taken at the United Nations Conference on the Human Environment held in Stockholm in June 1972, in which India participated, to take the appropriate steps for the preservation of the natural resources of the earth which, among other things, includes the preservation of the quality of air and control of air pollution. It was amended in the year 1987 (http://www.envfor.nic.in/legis/air/air1.html). The Central Pollution Control Board (CPCB) of India is a statutory organisation under the Ministry of Environment, Forest and Climate Change (MoEF&CC). It was established in 1974 under the Water Prevention and Control of Pollution Act. The CPCB is also entrusted with the powers and functions under The Air Prevention and Control of Pollution Act from 1981. The CPCB has set National Ambient Air Quality Standards for pollutants like $PM_{10}$, $PM_{2.5}$, $SO_2$, $NO_2$, $NH_3$, CO, BaP, and As. According to the standards given by the CPCB, the $PM_{10}$, $PM_{2.5}$, $SO_2$, $NO_2$, $NH_3$ 24 hours concentrations are 100 $\mu g/m^3$, 60 $\mu g/m^3$, 80 $\mu g/m^3$, 80 $\mu g/m^3$ and 400$\mu g/m^3$, respectively, for residential, industrial, rural and other areas; however, their annual concentrations are different. For the ecologically sensitive area, notified by Government of India, these standards vary (http://cpcb.nic.in/). Standards are also set for toxic elements like As, Pb, Ni, and BaP. The annual concentrations of these pollutants are 6 $ng/m^3$ for As, 20 $ng/m^3$ for Ni, and 1 $ng/m^3$ for PAHs.

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## SAŽETAK

### Usporedni pregled hrvatskih i indijskih znanstvenih istraživanja onečišćenja zraka s naglaskom na zagađivala povezana s izgaranjem ugljena

Zdravlje ljudi, okoliš i klima pod velikim su utjecajem složenih interakcija onečišćujućih tvari u atmosferi. Gotovo sve ljudske aktivnosti uzrokuju ispuštanje onečišćujućih tvari u zrak. Njihovo razumijevanje i kvantifikacija prvi je korak prema kontroli i ublažavanju onečišćenja zraka. Opći cilj ovoga rada jest prikaz rezultata odabranih hrvatskih i indijskih znanstvenih radova koji se bave onečišćenjem zraka, posebice onih s naglaskom na sumpor i aerosole povezane s elektranama na ugljen. Dvije zemlje u osnovi se razlikuju s obzirom na njihovu veličinu, zemljopisna obilježja, povijest, gospodarstvo i industrijski potencijal. Međutim, obje zemlje imaju određene veze s pridobivanjem fosilnih goriva i njihovom uporabom u elektranama i industriji već desetljećima. Prikazani su različiti znanstveni pristupi u istraživanjima te državne politike u legislativi onečišćenja zraka.

**Ključne riječi**
sumpor, aerosoli, izgaranje ugljena, onečišćenje zraka, istraživanja atmosfere