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Atmospheric Environment 40 (2006) 2046-2057



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Long-term trends of benzo(a)pyrene concentration on the eastern coast of the Baltic Sea

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Received 27 May 2005; received in revised form 9 November 2005; accepted 22 November 2005

Abstract

Concentration of benzo(a)pyrene (B(a)P) was investigated in atmospheric air at Preila background station located on the eastern coast of the Baltic Sea in 1980–2002. A significant difference in daily concentration of benzo(a)pyrene, reaching more than two orders of magnitude, was determined during the period of investigation. The variability of benzo(a)pyrene daily concentration was considered. The main part of extreme benzo(a)pyrene daily concentration of over 5 ng m^{-3} was related to the air masses coming to the background site from W-SW to N-NW over the period of 1980–1994. Owing to the variability of benzo(a)pyrene daily concentration, the monthly concentration of benzo(a)pyrene in atmospheric air of the background site varied from 0.18 to 3.30 ng m^{-3} in cold season and from 0.02 to 1.72 ng m^{-3} in warm season. A slowly decreasing trend of benzo(a)pyrene monthly concentration determined at Preila background station with that in other countries since 1994 (according to the EMEP program) has shown that monthly concentration of benzo(a)pyrene is nearly of the same level in the Czech Republic and it is by one order of magnitude higher than at Scandinavian background stations with tendency to decrease from 1999.

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Keywords: Benzo(a)pyrene; Concentration; Atmospheric air; Pollution sources

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) represent an important group of chemical pollutants, carcinogenic activity of which was investigated and confirmed at the very beginning of the last century. A different degree of carcinogenity of PAHs was determined. Among them benzo(a)pyrene was tested for nine animal species and was found to be the strongest carcinogen of this group of pollutants (Brookes, 1977; Griciute, 1979). Furthermore, the data of recent biomedical studies indicated that benzo(a)pyrene due to its chemical stability and lipophilicy can be accumulated in living tissues influencing transformation of living organism cells with genotoxic consequences (Binkova et al., 2003).

PAHs are produced by incomplete combustion processes of different organic fuel: wood, coal, oil, and its products (Lee et al., 1981). A significantly higher amount of carcinogen is emitted to the atmosphere by combustion of various sorts of coal than by other kinds of fuel (Brockhaus and Tomingas, 1976). Furthermore, it was determined

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 $^{1352\}text{-}2310/\$$ - see front matter \odot 2006 Elsevier Ltd. All rights reserved. doi:10.1016/j.atmosenv.2005.11.045

that emission from wood combustion was 400-fold larger than emission due to gasoline combustion and at least 10,000-fold larger than that from combustion of domestic heating oil (Harkov and Greenberg, 1985). A high amount of PAHs, partly benzo(a)pyrene, in the emissions of combustion processes allow using these pollutants as fuel combustion signatures for receptor modeling (Li and Kamens, 1993).

The variability of industrial objects, such as power stations, municipal incineration, petroleum refinery, domestic heating stoves and various types of vehicles, can be related to the generation of PAHs. In spite of carcinogenic risk and widely distributed sources of carcinogenic pollutants, the systematic investigations of PAHs were performed only at a few stations in Europe with predominant benzo(a)pyrene measurements. The first systematic measurements of benzo(a)pyrene were performed in eight towns of England in the fifties (Waller, 1952; Cooper, 1954), with further development of these investigations in other countries since 1960 (Sawicki et al., 1960; Monkman et al., 1962; Dikun, 1964; Grimmer, 1968; Bosco and Grella, 1966; Khesina, 1969; Shabad, 1973: Gordon, 1976: Brockhaus and Tomingas, 1976). Determination of benzo(a)pyrene at rural sites was started only in 1970 (De Wiest, 1978; Bjorseth et al., 1979; Broddin et al., 1980; Aleksejeva and Teplickaja, 1981; Milukaite, 1979, Milukaite and Galvonaite, 1986; Lyall et al., 1988; Rovinskij et al., 1988; Brorstrom-Lunden and Lovblad, 1991; Brorstrom-Lunden et al., 1994). Investigations of main PAHs concentrations in the environment and their sources as well as their transport over long distances have been dealt with international programs (EMEP, HELCOM, EUROTRAC) since 1990 (Pacyna et al., 1999; Pecar et al., 1999; Shatalov et al., 2001). The data of these programs were used in the adoption of Stockholm Convention on persistent organic pollutants (POPs) in 2001. As a result, the impact of these compounds on biota and humans has been studied (Holoubek et al., 2003). The measurements of PAHs and other POPs have been performed in the EMEP project since 1991 and data from a few stations are available via http://www.EMEP.int.

Recently, there is a lack of systematic long-term investigations of benzo(a)pyrene and only one publication covering a 12-year period, published by the scientist from EPA network in 1966–1977, is known (Faoro and Manning, 1981). It was shown that concentration of benzo(a)pyrene decreased by about four times from about 2.0 ng m^{-3} in 1967 to

 $0.5 \,\mathrm{ng}\,\mathrm{m}^{-3}$ in 1977 at nonurban sites due to the change-over of coal to oil or gas for heating in single and multiple family dwellings and other buildings. Different concentrations of benzo(a)pyrene with high fluctuations at a few stations and different time periods were determined in Europe during two last decades. A significant reduction of benzo(a)pyrene concentration from 46.0 ng m^{-3} in 1947–1951 to 26.0 ng m^{-3} in 1962–1963, to 5 ng m^{-3} in 1972–1973 and to 0.8 ng m^{-3} in 1991–1992 was determined in London for the 35-year period. A decrease in benzo(a)pyrene concentration by 1.5 orders of magnitude at urban sites caused of benzo(a)pyrene concentration decrease by 2-4 times in rural atmospheric air of UK during 1960s and 1990s (Jones et al., 1989; Halsall et al., 1994). The daily concentration of benzo(a)pyrene varied in the range of $2-202 \text{ ng m}^{-3}$ at three Belgium rural sites in wintertime in 1975–1976, with transport of highly polluted air masses from Great Britain and Ruhr area (De Wiest, 1978). Still high concentration of benzo(a)pyrene in atmospheric air of rural sites was determined in upper Silesia, Poland, during the 1988-1989 period (Bodzek et al., 1993). Concentration of benzo(a)pyrene in atmospheric air at nine sites varied between 8.7 and 41.6 ng m^{-3} in summertime. At almost all sites of sampling the standart limit of benzo(a)pyrene concentration in the air (10 ng m^{-3}) was exceeded. This was justified by high annual emission from numerous industrial plants in Silesia region. Long-term investigations of benzo(a)pyrene at Lithuanian rural site showed that daily concentration of carcinogen can vary between 0.02 and 25.08 ng m^{-3} in the period of 1980–1994 (Milukaite et al., 1995). Two-fold lower concentration of benzo(a)pyrene in atmospheric air was determined at Rucava background station as compared to that at Preila background station, although the stations are at a distance of 100 km and analysis of benzo(a)pyrene was performed in one and the same laboratory. Differences were justified not only by various local sources but by different frequency of south-western air masses at these neighbour background stations (Milukaite et al., 2004). The measurements in northern Bohemia (the Czech Republic) show that the highest average wintertime concentration of benzo(a)pyrene reaching over 5 ng m⁻³ in 1995–1997 has decreased about two times since 1998 (Binkova et al., 2003). Relatively low concentration of BP was determined at the Košetice background station located in the southern part of Czech Republic in 1994-1995,

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