ABSTRACT

Concentrations of polycyclic aromatic hydrocarbons (PAHs) were measured in sediment cores collected from 11 inland lakes in Michigan, for examination of spatial and temporal trends in accumulation and sources. Concentrations of PAHs varied widely among these lakes; the highest concentrations were found in sediment collected from Cass Lake, located in an urbanized watershed. Concentrations of PAHs were correlated with watershed population density, on a spatial scale. PAH concentrations have decreased over time since the 1980s in nine of the 11 lakes studied, most of which are located in the suburban and rural areas. The rate of decline in PAH concentration was faster in Gratiot Lake, a remote lake with no known local sources of pollution in recent years. Concentrations of PAHs in surface sediments from Cass and Cadillac Lakes were greater than the threshold for adverse effects. Despite the wide variations in total PAH concentrations, fluoranthene, pyrene, chrysene, benzo[b]fluoranthene and benzo[k]fluoranthene were the major PAHs in all of the lakes studied. The proportions of individual PAHs within the total PAH concentrations have changed both temporally and spatially, suggesting a shift in the sources of PAHs in the environment. A general increase in the proportion of high-molecular-weight PAHs suggests an increase in combustion-related sources.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants, released into the environment primarily by incomplete combustion of fossil fuels and burning of vegetation and other organic materials. This study compares spatio-temporal patterns of PAH accumulation among a number of inland lakes to determine current and historical influences of PAH loadings to the Great Lakes region.

MATERIALS & METHODS

Sediment cores were collected from Cass, Elk, Gratiot, Gull, and Higgins Lakes in the summer of 1999 (Figure 1). Sediment cores were collected from Crystal and Littlefield Lakes in the summer of 2000. Sediment cores from Cadillac, Crystal, Mullett, and Whitmore Lakes were collected in 2001. The Crystal Lakes from Montcalm and Benzie Counties are referred hereafter as Crystal M and Crystal B, respectively. Lakes were also chosen to represent a broad range of human disturbance, from areas of high development (e.g., Cass Lake) to relatively remote lakes (e.g., Gratiot Lake).

PAH Concentrations. Concentrations of total PAHs (sum of 16 priority PAHs) and selected individual PAHs in surface (0-2 cm) sediments from all of the lakes studied are shown in Table 1. PAH concentrations varied considerably among the lakes. A maximum PAH concentration of 16800 ng/g, dry wt, found in the surface sediment from Cass Lake, was the highest concentration measured in this study. High concentrations of PAHs in Cass and Cadillac Lakes can be related to the high population densities of these two lakes’ watersheds.

Watershed population densities of all of the lakes in 2000 were plotted against total PAH concentrations measured in surface sediments (Figure 2). In general, the concentration of total PAHs correlated significantly with watershed population density, for each lake ($r^2=0.66; P<0.05$).

RESULTS & DISCUSSION

Overall, concentrations of PAHs in inland lakes increased from the early to mid 1900s onwards, and then declined after the 1980s (Figs.3 and 4). For instance, the overall rate of decline in PAH concentrations from 1985 to 2000 ranged from 19 to 85% (for 15 years) for all of the lakes, except Cass and Higgins Lakes, which recorded an increase in PAH concentrations recently. Urban sprawl, with concomitant increase in automobile use, was suggested as an explanation for the recent increase in PAH concentrations in urban lakes. The gradual decrease in PAH concentrations in suburban and rural lakes is consistent with the shift to alternative energy sources in industrial processes, and implementation of particulate emission controls, have reduced regional-scale emissions of PAHs.

PAH Profiles and Sources

Four- and five-ring PAHs were the most abundant compounds in sediments, whereas two- and three-ring PAHs were less abundant (Fig.5). The differences in proportions of individual PAHs indicate the influence of local sources or processes in each of the lakes. Temporally, it is worth noting that the types of sources of PAHs in these lakes have shifted.

Studies have shown that coal/coking-related sources of PAHs have declined recently in areas distant from steel production, while PAHs emitted from motor vehicles have increased. The relative increase in the proportion of high-molecular-weight PAHs suggests a general shift in PAH sources. Uncombusted sources (e.g., oilseeds, petroleum spills) contain predominantly two- and three-ring PAHs, whereas combustion sources (e.g., vehicle exhaust, domestic heating with coal, forest fires) contain predominantly four- and five-ring PAH species. A general increase in the proportion of high-molecular-weight PAHs suggests an increase in combustion-related sources in recent years.

Temporal Trends

Fig. 1. Lakes sampled in the summers of 1999-2001.

Fig. 2. Relationship between watershed population density (in 2000) and PAH concentration in surface sediments from several inland lakes in Michigan.

Fig. 3 (left) and Fig 4 (right). Temporal trends in PAH concentrations in Michigan lakes.

Table 1. Concentrations of PAHs (sum of 16 priority PAHs) and selected individual PAHs in surface sediments (0-2 cm) from Inland Lakes in Michigan.