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Atmospheric particulate polycyclic aromatic hydrocarbons from road transport in southeast Brazil

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ABSTRACT

The Cubatão industrial complex of southeast Brazil is sited on a coastal strip bounded on the continental side by a mountainous scarp covered with tropical forest. Four cities have developed around the complex. The metropolitan region of São Paulo is located on a plateau above the scarp. The combination of the region's topography, the industrial installations, and an intensely trafficked road network causes widespread atmospheric pollution. In November 2004, measurements were made of the 16 polycyclic aromatic hydrocarbons designated as priority pollutants by the US Environmental Protection Agency, as well as of aerosol mass and ionic composition. Use of characteristic concentration ratios for emission sources show that tailpipe emissions from diesel vehicles was the main source of the compounds. This means that a shift from gasoline to ethanol as fuel in spark ignition engines will have only minor influence on atmospheric polycyclic aromatic hydrocarbon concentrations, despite very low emissions during ethanol combustion. On the other hand, reduction in emissions associated with increasing use of biodiesel in compression ignition engine fuel mixtures could significantly reduce atmospheric concentrations.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAH) are released into the environment from anthropogenic and natural sources, and are of concern due to their toxicity to humans. Incomplete combustion of transport fuels, biomass, coal and oil are the main sources, plus processing of fossil fuels, metal smelting and coke production, and manufacture and use of preservatives (UN Environment Programme, 1998, and references therein; Finlayson-Pitts and Pitts, 2000). Natural sources of PAH include coal and crude oil, volcanoes (Ilnitsky et al., 1977) and wildfires (Baek et al., 1991). PAH are not only toxic in their native forms, but can also produce carcinogenic products (for example nitro-PAH) following reactions with other atmospheric species such as nitrogen oxides.

In the atmosphere, phase partitioning of PAH between particles and gases depends on compound volatility, which decreases with increasing molecular weight, and other factors such as atmospheric aerosol concentrations (Baek et al., 1991). The compounds are partially soluble in water, with solubility decreasing with increasing molecular weight (Mackay et al., 1992). From a human health perspective, the particle phase is of greatest concern, due to the greater likelihood of deposition in the deeper respiratory system, following inhalation (Kameda et al., 2005). Condensed PAH released during combustion are present in very small particles (<0.5 μm aerodynamic diameter), however the size distribution changes

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during atmospheric transport. As a result, higher molecular weight compounds are normally found within small particles (<2 μm), with volatilisation and re-condensation causing transfer of semivolatile species from smaller to larger particles, and development of a size mode around 0.5–4.0 μm (Miguel et al., 2004). Most reported measurements have focussed on the 16 PAH identified by the US Environmental Protection Agency as priority compounds, which have molecular weights of between 128 Da and 276 Da. Although larger PAH having molecular weights greater than 300 Da may be present, these have not been routinely measured, due to analytical difficulties.

We report the first measurements of priority particulate PAH in the coastal atmosphere of southeast Brazil, a densely populated region where there is intense heavy-duty goods vehicle movement due to the proximity of one of the Southern Hemisphere's most concentrated installations of heavy industry (the Cubatão industrial park) as well as South America's largest commercial port (Santos). Air quality in the region is severely compromised by pollutants emitted from transport and industrial sources (Klockow and Targa, 1998).

2. Experimental

Aerosols were sampled onto quartz fibre filters, over periods of 24 h, using a high volume sampler (Thermo Andersen, USA) configured to collect particles possessing aerodynamic diameters of 10 μm or less (PM_{10}). These measurements were made during 24th–28th and 29th–30th November 2004, in the Baixada Santista air basin (23°53″S, 46°25″W), near the town of Cubatão with its adjacent industrial park (Fig. 1). $PM_{2.5}$ (particles possessing aerodynamic diameters of 2.5 μm or less) and PM_{10} samples were also collected using low volume samplers, for analysis of soluble ions.

Filters were weighed before and after sampling under conditions of controlled temperature and humidity, in accordance with the International Organization for Standardization procedure (No. 15767). Extraction and analysis of the PAH from filters was according to previously published methods (Lopes et al., 2008, and references therein). Analytical standard solutions were obtained from AccuStandard Inc., USA. Recoveries, determined as the averages of three extractions of standard reference material (SRM 1649, urban dust, organics, National Institute of Standards and Technology, Washington, DC), were between 45% (fluorene) and 107% (dibenz[*a*,*h*]anthracene), with an overall mean of 80%. Anions and cations in aqueous filter extracts were determined using ion chromatography (Allen et al., 2004).

3. Results and discussion

3.1. Meteorology

Diurnal changes in wind direction occur due to the geographical location on a coastal plain at the base of a 900 m coastal mountain range (the Serra do Mar), with day time sea breeze development between the hours of ca. 11:00 am and 20:00 pm, and down-slope (katabatic) air flows at night. Maximum (day time) and minimum (night time) temperatures were 27–30 °C and 19–23 °C, respectively. Relative humidity was 51–85% during the day (100% after November 28th due to persistent rain), and 100% at night. At night, under stable conditions, down-slope flows (wind speeds 0.9–2.3 m s^{-1}) brought cleaner air

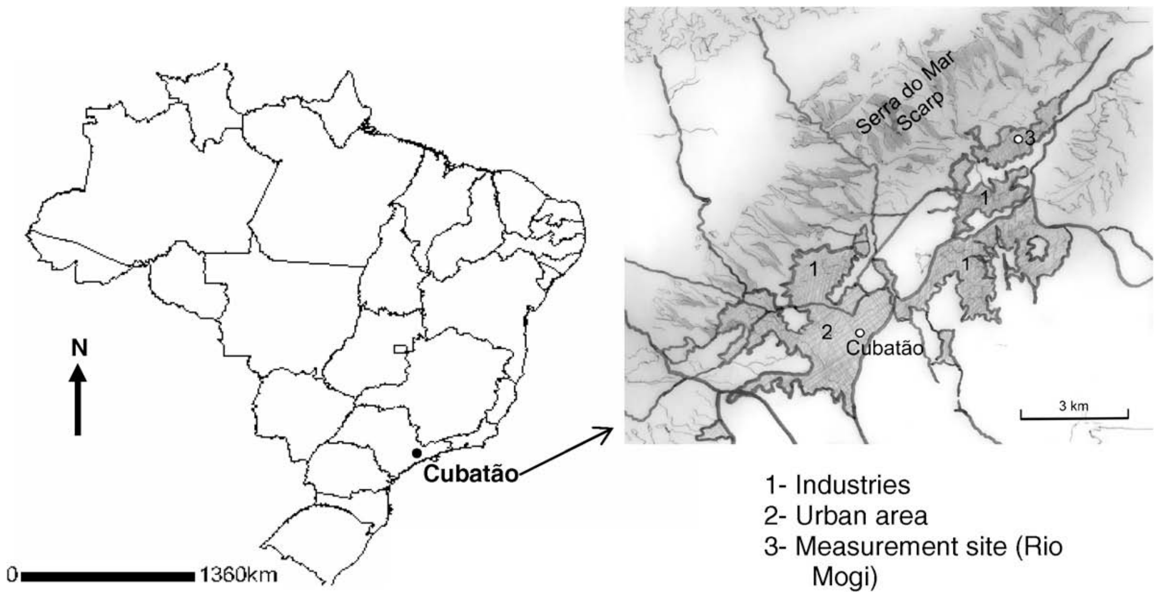

Fig. 1. Map of Cubatão, and surrounding region, showing location of the Rio Mogi site.

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