



## Air–water exchange fluxes of polycyclic aromatic hydrocarbons in the tropical coast, Taiwan

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### HIGHLIGHTS

- ▶ PAH distribution in the air and water of the tropical, rural coast was studied.
- ▶ Sensitive research-grade sampling and analytical method with a high-QCQA was used.
- ▶ PAH flux across air and water was influenced by its dissolved level and wind speed.
- ▶ Annual flux of PAHs were measured and compared with other studies.

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### ABSTRACT

Air–water exchange fluxes of polycyclic aromatic hydrocarbons (PAHs) were simultaneously measured in air and water samples from two sites on the Kenting coast, located at the southern tip of Taiwan, from January to December 2010. There was no significant difference in the total PAH (t-PAH) concentrations in both gas and dissolved phases between these two sites due to the less local input which also coincided to the low levels of t-PAH concentration; the gas and dissolved phases averaged  $1.29 \pm 0.59 \text{ ng m}^{-3}$  and  $2.17 \pm 1.19 \text{ ng L}^{-1}$  respectively. The direction and magnitude of the daily flux of PAHs were significantly influenced by wind speed and dissolved PAH concentrations. Individual PAH flux ranged from  $627 \text{ ng m}^{-2} \text{ d}^{-1}$  volatilization of phenanthrene during the rainy season with storm–water discharges raising dissolved phase concentration, to  $67 \text{ ng m}^{-2} \text{ d}^{-1}$  absorption of fluoranthene during high wind speed periods. Due to PAH annual fluxes through air–water exchange, Kenting seawater is a source of low molecular weight PAHs and a reservoir of high molecular weight PAHs. Estimated annual volatilization fluxes ranged from  $7.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$  for pyrene to  $50 \mu\text{g m}^{-2} \text{ yr}^{-1}$  for phenanthrene and the absorption fluxes ranged from  $-2.6 \mu\text{g m}^{-2} \text{ yr}^{-1}$  for chrysene to  $-3.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$  for fluoranthene.

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### 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of ubiquitous environmental persistent organic pollutants (POPs) containing two or more fused aromatic rings made up of carbon and hydrogen atoms, and have been recognized as a major environmental and health concern due to their potential accumulation and mutagenic and carcinogenic effects on organisms (Neff, 2002). PAHs are mainly produced by the incomplete combustion of organic material and fossil fuels which results from industrial

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processes and other human activities. The report of the global atmospheric emission inventory of PAHs displayed that biofuel (57%), wildfires (17%) and consumer product usage (7%) were the major sources of PAHs, and that China, India and United States were the three countries with the highest PAH emissions (Zhang and Tao, 2009). PAHs are semi-volatile organic compounds (SVOCs) with vapor pressures in the range of  $10^{-4}$ – $10^{-11}$  atm, and they are transferred throughout the world via successive deposition and re-emission (Ottar, 1981). In fact, PAHs are mainly associated with aerosol soot carbon, which make high MW PAHs poorly available in the dissolved phase (Dachs and Eisenreich, 2000). Also, the partitioning of PAHs in the atmosphere affects their transport fates. Many studies have pointed out that ambient temperature is highly correlated with gas–particle partitioning of SVOCs (Pankow and Bidleman, 1991; Chen and Preston, 1997).

PAHs are present in the atmosphere both in the gas phase and associated with particles which may be transferred long distance from remote regions (Lohmann et al., 2000; Mader and Pankow, 2002). Chemical loading of PAHs into the aquatic system may come from precipitation scavenging, dry deposition or air–water exchange. The gas exchange across the air–water interface is one of the major processes that control the concentrations and residence times of PAHs in natural water bodies. PAHs of high molecular weight (HMW), mostly exclusively associated with aerosols, exist in the atmosphere and enter the surface water through dry deposition and aerosol scavenging by precipitation. Once deposited, particulate PAHs may not be rapidly equilibrated with the dissolved phase and are not volatile in the short term. Slow release of HMW PAHs from particles may result from slow desorption kinetics and diffusion from the interior of the aquatic particles. Conversely, lower molecular weight PAHs exist in both gas and aqueous dissolved phases, and are available for active gas exchange across the air–water interface. The rate of diffusive exchange across the air–water interface may be modeled as a first-order mass transfer process if the compound mass transfer coefficients for specific environmental conditions are known. Thus, simultaneous sampling of air and surface seawater is necessary to describe the mass transfer of contaminants by gas exchange across the air–water interface according to the meteorological and geographical conditions (Bidleman, 1999).

The dynamics of air–water exchange and processes within the ocean are critical to the global fate and behavior of POPs. The capacity of surface waters to store POPs that are spatially and temporarily available is influenced by the temperature, mixing depth, and biogeochemical processes (Jaward et al., 2004). The magnitude and direction of air–water exchange have been investigated extensively because of its significant contribution to the cycling of POPs in the environment. Paired air and water concentration measurements used in conjunction with the two-layer film model have been commonly used to estimate atmospheric loading of POPs to water bodies or their loss by volatilization (Hoff et al., 1996; Nelson et al., 1998; Bamford et al., 2002; Gigliotti et al., 2002). However, the air–water exchange of PAHs in southern Taiwan has not been investigated using paired samples.

The Kenting coast is located at the southernmost tip of Taiwan, adjacent to the South China Sea. The beautiful landscape of the Kenting attracts millions of tourists annually, and the increasing tourist activities threaten the marine environmental of the Kenting area. The concentrations of PAHs in the coastal seawater and sediment have been measured discretely in our previous studies (Cheng et al., 2010, 2012) but neither considered the interaction between the atmosphere and water. The flux of PAHs may be related to high temperature and solar intensity (Galarneau, 2008). The Kenting coast is located in a transitional zone of the East Asian monsoon system, which includes the moist southwestern wind from the South China Sea during the summer, and dry northeastern wind (with extremely high-speed) from mainland China during the winter. The dynamic geochemical cycle of Kenting seawater is driven by summer typhoons, river input, plankton blooms, summer stratifications, winter water column disturbance by the high wind speeds, and tide-induced upwelling (Chen et al., 2004; Lee et al., 1997, 1999; Meng et al., 2008). Due to the dynamic wind directions and geochemical cycles of water bodies, different atmospheric contaminants have different concentrations and fates via the air–water exchange on the Kenting coast. The objectives of this study were to quantify the magnitude and direction of the air–water exchange, to estimate the annual fluxes, and to investigate the dynamic air–water coupling of PAHs by simultaneously measuring the gaseous and dissolved PAH concentrations in the Kenting coastal area.

## 2. Materials and methods

### 2.1. Sampling sites and meteorological conditions

The water samples were collected from two sites on the Kenting coast (Fig. 1). Hunwan station (HW, 22°02'46" N, 120°42'9" E), located in west Kenting, is influenced by continental runoff from Si-chon River and Baoli River outflows. Nanwan station (NW, 21°56'56"N, 120°46'51"E), located at the southern end of Kenting, has relatively higher levels of human activity and traffic, and is influenced by continental runoff from Shiniu River outflow, waste water treatment plants and maritime harbor activity. The air samplers were placed near each water sampling station (less than 1000 m) at the National Museum of Marine Biology and Aquarium (NMMBA) and Kenting National Park (KTNP).

Atmospheric data were downloaded and obtained from the [Data Bank for Atmospheric Research](#). Meteorological data (air temperature, wind speed and direction, precipitation, and relative humidity), were obtained from the meteorological station (Central Weather Bureau in Hengchun; Fig. 1) located between these two sampling sites and surface water temperature and salinity were measured on-site using a portable apparatus (YSI Model 85, USA).

### 2.2. Sample collection

The air and water samples from both the paired-sites of NMMBA-HW and KTNP-NW were taken concurrently every two weeks from January to December 2010; summarily, 21 paired samples were collected from each site. The air samples were collected with a high volume air sampler (TE-1000 PUF, Tisch Environmental, USA), with flow rate set to 0.195 m<sup>3</sup> min<sup>-1</sup> and run for 48 h. After the air samples were pumped through the quartz fiber filter (Tissuquartz, 2500QAT-UP, 102 mm, PALL), the gas phase was captured on polyurethane foam (PUF; 3 cm × 5 cm). The PUF plugs were cleaned in a Soxhlet apparatus re-fluxed with petroleum ether for 24 h, then dried in the oven at 50 °C. Before field sampling, the PUF was installed in a solvent-rinsed glass cylinder and carefully sealed with aluminum foil. The flow rate of the air sampler was re-calibrated every 3 months. Air volumes collected were nominalized by m<sup>3</sup>, based on the average of the initial and final air velocities for each sample.

While the air samples were pumped, surface water samples from HW and NW stations were collected in two 20 L polished stainless steel cans (total volume 40 L). The collected seawater samples were pushed by a pressurized nitrogen stream through a 293 mm diameter GFF filter (mesh = 1 μm) placed inside an aluminum filter holder to collect and remove the aqueous particles. The filtered water was then passed through a glass column (35 cm × 1.5 cm) packed with Amberlite XAD-2 resin to capture the dissolved phase PAHs. Before use, the Amberlite XAD-2 resin was Soxhlet-extracted with methanol, acetone, hexane, dichloromethane, hexane, acetone, and then methanol separately for 24 h each and then rinsed with deionized (Milli-Q) water. Cleaned resin was kept in sealed jars with Milli-Q water and loaded in glass columns prior to sampling. The exit flow rate of the water sample from the glass column was maintained at 250–300 ml min<sup>-1</sup> by pressurized nitrogen.

### 2.3. Extraction and clean up

The XAD-2 resin was extracted in a Soxhlet apparatus for 24 h with acetone and *n*-hexane (1:1). The extract was washed in a separatory funnel with 50 mL Milli-Q water three times. The resulting aqueous phase was further extracted with 10 mL *n*-hexane. The extract was then concentrated to about 5 mL by

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