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著者	Asano Kohei, Kameda Takayuki, Sadanaga Hiroyasu, Takenaka Norimichi, Bandow Hiroshi
journal or publication title	Promotion Environmental Research in Pan-Japan Sea Area -Young Researchers' Network- : Abstract
page range	147-148
year	2006-03-08
URL	http://hdl.handle.net/2297/6572

Study on formation processes of nitrated polycyclic aromatic hydrocarbons in the atmosphere

Kohei ASANO^a, Takayuki KAMEDA^b, Hiroyasu SADANAGA,

Norimichi TAKENAKA^a, and Hiroshi BANDOW^a

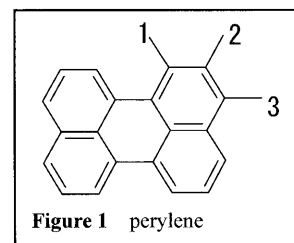
(a) Graduate school of engineering, Osaka Prefecture University, 1-1, Gakuen-cho, Sakai, Osaka, 599-8531, JAPAN

(b) Institute of Nature and Environmental Technology, Kanazawa University, Kakuma-machi, Kanazawa, Ishikawa, 920-1192, JAPAN

Introduction

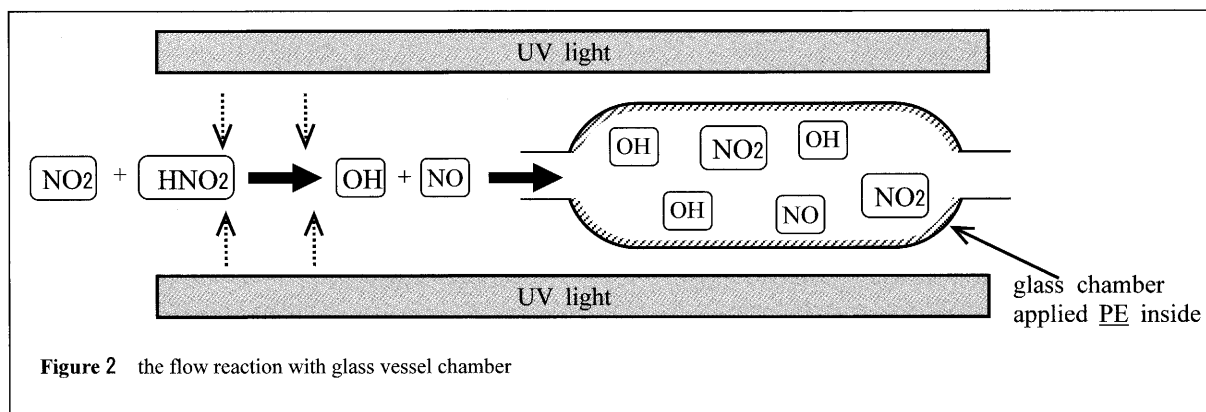
Nitrated polycyclic aromatic hydrocarbons (NPAHs), most of which have strong mutagenicity, are detected in the atmosphere. Nevertheless, the formation process of NPAHs is still uncertain. In our previous study, reactions between atmospheric radicals (i.e. OH and NO₃) and polycyclic aromatic hydrocarbons (PAHs) in the gas phase have been investigated with an assumption that some of the NPAHs could be yielded through an atmospheric reaction of parent PAHs. However, whole list of the NPAHs observed in ambient air was not covered on the products of the experiments. In this study, heterogeneous reactions between radicals and PAHs on the particle were studied as another production process of NPAHs in the ambient air.

Perylene (PE) has been found on the particle in ambient air. PE has very low volatility, and thus it could be rational to assume that homogeneous reaction of PE with atmospheric radicals can be neglected. From this point of view, PE was selected as an objective substance in this study.



Experimental

The flow reaction was conducted on the glass vessel chamber (5 cm i.d. × 18 cm). PE/benzene solution (1 mM, 5 ml) was put on the glass vessel chamber, and evaporated with N₂ gas. Then, PE was exposed by HNO₂ gas under the UV light irradiation, HNO₂ gas played the role of the source of OH radicals (HNO₂ + hv (354 nm) → NO + OH).



^a Electronic Address: kohei-a@chem.osakafu-u.ac.jp URL: <http://www.ams.osakafu-u.ac.jp/lab2/home/>

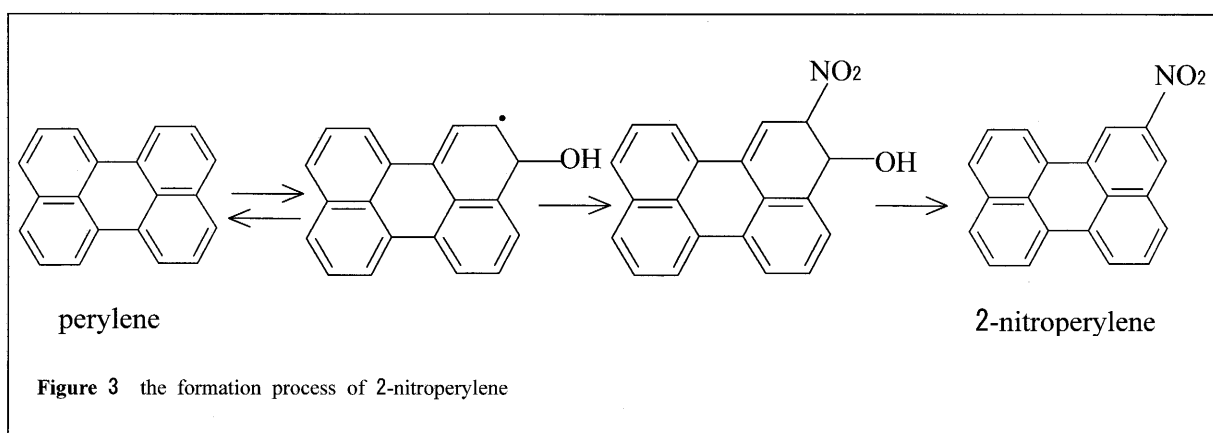
HNO₂ was generated by the reaction of sodium nitrite with gaseous hydrochloric acid,¹⁾ (NaNO₂ + HCl → NaCl + HNO₂).

HNO₂ concentration thus generated was *ca.* 1.5 ppmv, and *ca.* 100 ppbv of NO₂ co-existed as a byproduct from the HNO₂ generation reaction. Therefore, another addition of NO₂ into the reaction system was not needed to form nitroperylene (NPE).

Reactants were extracted with 10 ml of benzene/ethanol (3/1, v/v) and analyzed by electron impact (EI) gas chromatography – mass spectrometry (GC-MS, Hewlett-Packard). An HP-1MS column [0.25 mm i.d. × 15 m, 0.25 μm film thickness] was employed with a temperature program as follows: initial temperature at 50 °C, then temperature-programmed to 300 °C at 20 °C min⁻¹.

Results and Discussion

NPE has three isomers such as 1-NPE, 2-NPE, and 3-NPE. If PE reacted with OH radicals on the glass vessel chamber, the reactants should be 2-NPE. The formation process of NPE from the OH radical-initiate reaction can be thought as follows; at first an OH radical should attack the position 3 because PE has the highest electron density in there. Subsequently, a NO₂ attacks the position 2, finally the OH radical desorbs. 2-nitrofluoranthene, one of the famous NPAHs, is formed through the same scheme.²⁾



The reduction of PE concentrations was confirmed, and the results that suggest the appearance of NPE were obtained, but it has not identified yet through lack of the 2-NPE standard substances.

We will show more minute results from the analysis with the standard substances of 1-NPE and 3-NPE in the symposium.

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