Supplementary Information

Title: Polycyclic aromatic hydrocarbons (PAHs) and their alkylated (RPAHs), nitrated (NPAHs) and oxygenated (OPAHs) derivatives in the global marine atmosphere: occurrence, spatial variations, and source apportionment

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Text S1 Instrumental analysis

A gas chromatograph (Agilent 7890B GC) coupled with the mass spectrometer (Agilent 5977B MS) was employed to determine the concentrations of PAH, NPAH, and OPAH species. Capillary column DB-5MS was used to analyze PAH concentrations. The gas chromatograph oven temperature was programmed to kept at 70 °C for 1 min, to 260 °C at the rate of 10 °C/min, and then increased to 300 °C at the rate of 5 °C/min and remained the same temperature for 5 min. The carrier gas is high purity helium.

Both of NPAHs and OPAHs concentrations were determined using a DB-5MS capillary column in a negative chemical ionization mode. The gas chromatograph oven temperature was programmed to increase from 40 °C to 150 °C at the rate of 15 °C/min, to 300 °C at the rate of 5 °C/min, and then kept at the same temperature for 15 min. The carrier gas and reagent gas were high purity helium and high purity methane, respectively.

RPAH was analyzed using a GC (7890N GC, Agilent, Santa Clara, USA) equipped with a deactivated fused silica guard column (5 m, Agilent, Santa Clara, USA) followed by a 30 m × 0.25 mm × 0.25 μm fused silica column (DB-5MS, J&W, Santa Clara, USA). The gas chromatograph oven program was set to 80 °C as the initial temperature, and programmed to increase at a rate of 21°C/min to 150 °C. Then, the temperature was further programmed to increase at the rate of 5 °C/min to 300 °C. Finally, the air temperature kept at 300 °C for 20 min.

Table S1 Limits of detection (LOD) and recoveries of parent PAHs and derivatives.

Compounds	Compound class	LOD (ng/m^3)	Recovery (%)
Nap	PAHs	0.10	81
Acy	PAHs	0.09	80
Ace	PAHs	0.10	82
Flu	PAHs	0.13	86
Phe	PAHs	0.11	80
Ant	PAHs	0.02	80
Fluoran	PAHs	0.03	85
Pyr	PAHs	0.10	84
BaA	PAHs	0.04	83
CT	PAHs	0.25	85
BbF	PAHs	0.34	89
BkF	PAHs	0.11	93
BeP	PAHs	0.28	92
BaP	PAHs	0.09	95
Per	PAHs	0.02	83
IP	PAHs	0.14	86
BghiP	PAHs	0.02	88
DBA	PAHs	0.10	89
1,4-Naphthoquinone	OPAHs	0.01	85
1-Naphthaldehyd	OPAHs	0.03	81
1-Ancenaphthenaquinone	OPAHs	0.01	91
9-Fluorenone	OPAHs	0.01	80
Ancenaphthenaquinone	OPAHs	0.03	88
Anthraquinone	OPAHs	0.01	97
Benzanthrone	OPAHs	0.01	98
Benzo(a)anthracene-7,12-dione	OPAHs	0.02	119
1,4-Chysenequione	OPAHs	0.37	133
5,12-Naphthacenequione	OPAHs	0.04	113
6H-Benzo(cd)pyrene-6-one	OPAHs	0.02	137
2-Methylnaphthalene	RPAHs	0.01	86
1-Methylnaphthalene	RPAHs	0.02	88
2,6-Dimethylnaphthalene	RPAHs	0.02	85
1,3-Dimethylnaphthalene	RPAHs	0.03	92
1,6,7-Trimethylnaphthalene	RPAHs	0.02	83
2-Methylanthracene	RPAHs	0.01	82
1-Methylphenanthrene	RPAHs	0.01	89
1-methylanthracene	RPAHs	0.01	92
1-Methylfluoranthene	RPAHs	0.04	91
1-Nitronaphthalene	NPAHs	0.02	87
2-Nitronaphthalene	NPAHs	0.03	83
1,3-Dinitronaphthalene	NPAHs	0.03	82
5-Nitroacenaphthene	NPAHs	0.01	94

2-Nitrofluorene	NPAHs	0.04	95
9-Nitroanthracene	NPAHs	0.01	92
1,8-Dinitronaphthalene	NPAHs	0.01	90

Figure S1 The spatial distributions of OPAH species in the global marine aerosols (Unit: pg/m^3).

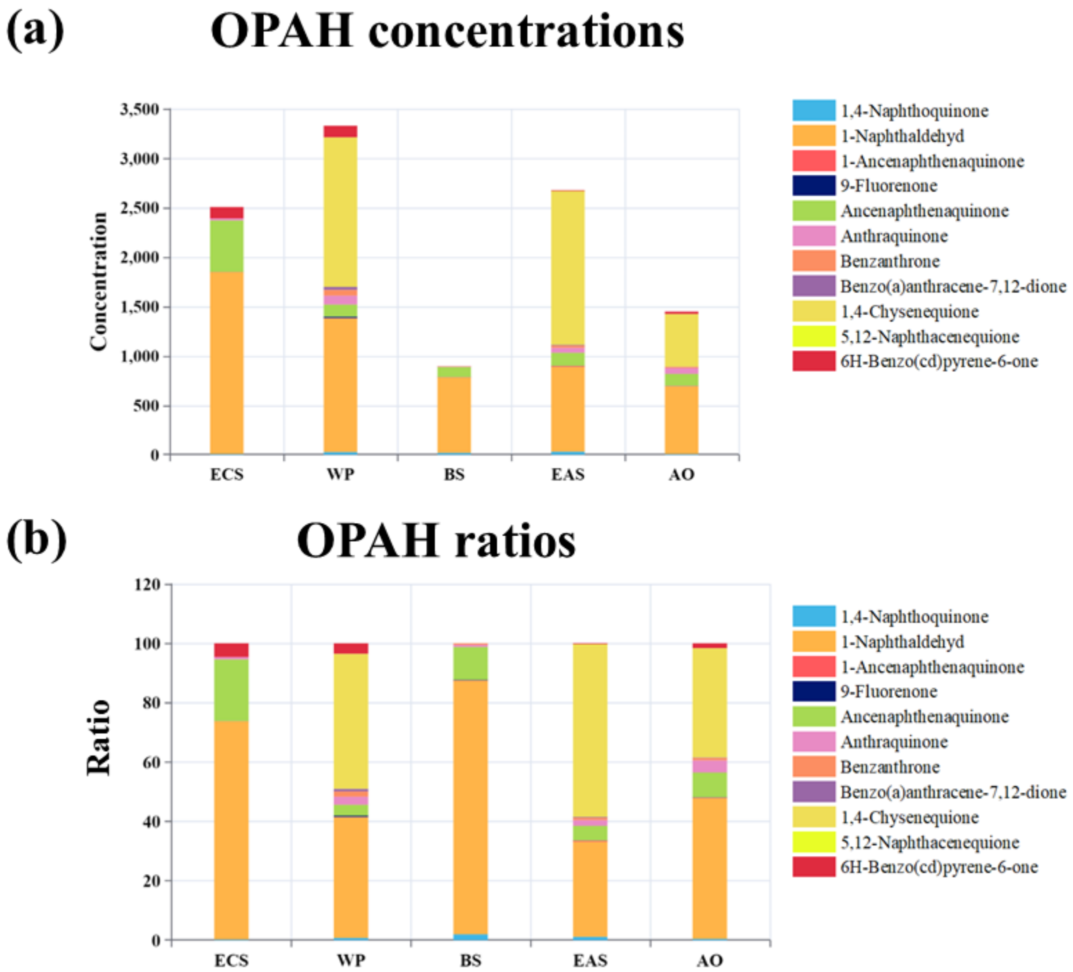

Figure S2 Global simulated wildfire-related benzene concentrations in 2019 based on GEOS-Chem model (Unit: ppb).

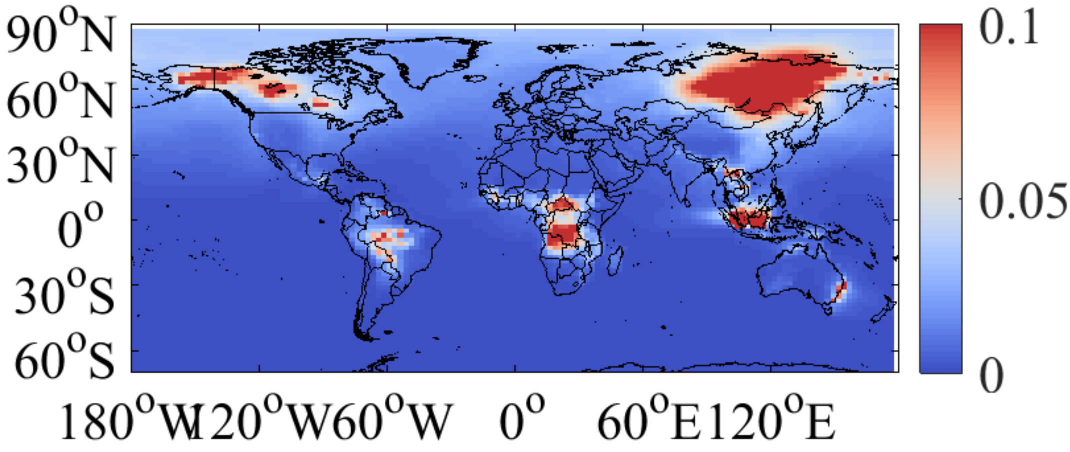

Figure S3 The global simulated ambient concentrations of O_3 (Unit: μg/m^3) and OH radical (Unit: 10^{-3} ppt) in 2019.

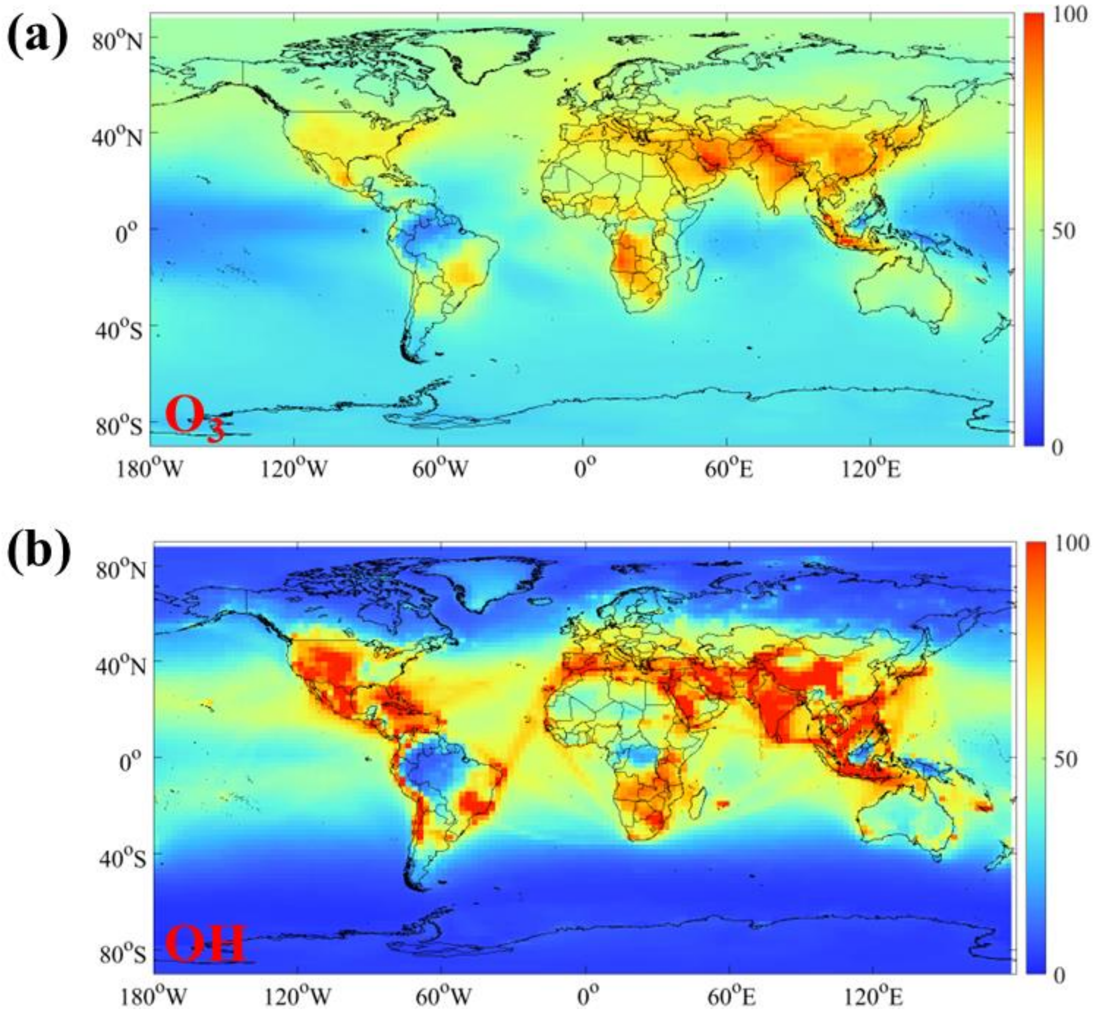

Figure S4 The spatial distributions of RPAH species in the global marine aerosols (Unit: pg/m^3).

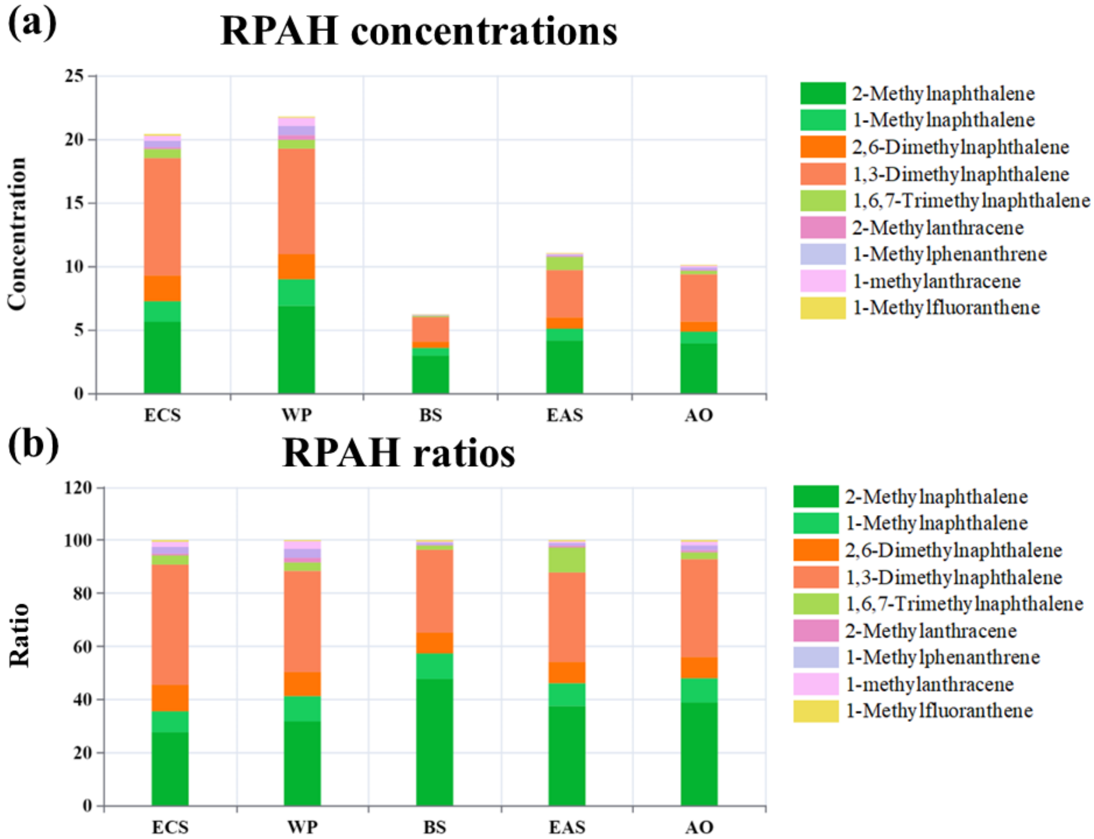

Figure S5 The spatial distributions of NPAH species in the global marine aerosols (Unit: pg/m^3).