**Supplementary Materials**

# Seasonal variability and risk assessment of atmospheric polycyclic aromatic hydrocarbons and hydroxylated polycyclic aromatic hydrocarbons in Kanazawa, Japan

**Masato Honda [1,*], Kazuichi Hayakawa [2], Lulu Zhang [2,3], Ning Tang [2] and Hiroyuki Nakamura [4]**

1 Botanical Garden, Institute of Nature and Environmental Technology, Kanazawa University, Kakuma, Kan-azawa, Ishikawa 920-1192, Japan

2 Institute of Nature and Environmental Technology, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan

3 School of Civil Engineering, Architecture and Environment, Hubei University of Technology, Wuhan 430068, China

4 Department of Hygiene and Public Health, Faculty of Medicine, Institute of Medical, Pharmaceutical and Health Sciences, Kanazawa University, Kanazawa City 920-8640, Japan

*Corresponding author: Masato Honda

Botanical Garden, Institute of Nature and Environmental Technology, Kanazawa University, Kakuma, Kanazawa, Ishikawa 920-1192, Japan

Tel: +81-076-264-5874

E-mail: mst-honda@se.kanazawa-u.ac.jp

**For submission to: Applied Sciendes, Special Issue "$PM_{2.5}$ and $PM_{10}$: Atmospheric Behaviors and Correlation with Health Effects"**

**Figure S1.** Box plot of seasonal atmospheric concentrations of PAHs in Kanazawa.

**Figure S2**. Box plot of seasonal atmospheric concentrations of OH-PAHs in Kanazawa.

**Figure S3**. PCA score plot in which the first two PCs for atmospheric PAHs.

**Figure S4**. PCA score plot in which the first two PCs for atmospheric OH-PAHs.

**Figure S5**. Diagnostic ratios calculated for atmospheric PAHs

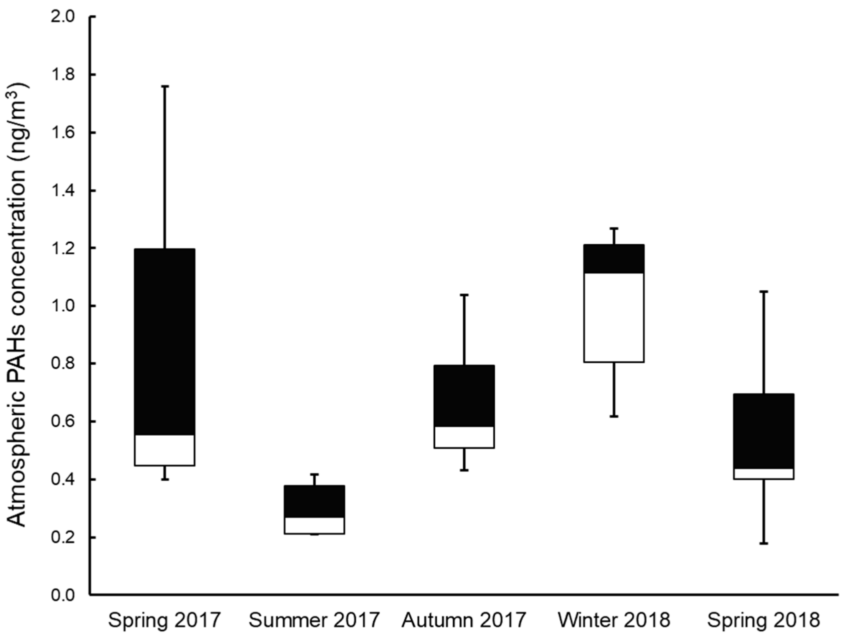
**Table S1.** Estrogenic and antiestrogenic activities of OH-PAHs from a yeast two-hybrid assay conducted by Hayakawa et al. (2007) [1].

**Table S2.** Binding affinities of PAHs and OH-PAHs to hER conducted by Hayakawa et al. (2007) [1].

## Reference

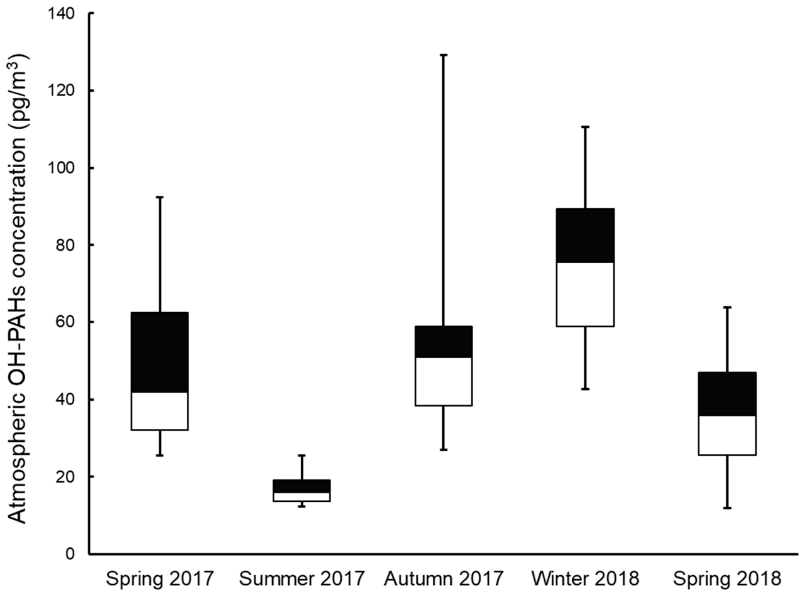

1. Hayakawa, K.; Onoda, Y.; Tachikawa, C.; Hosoi, S.; Yoshita, M.; Chung, S.W.; Kizu, R.; Toriba, A.; Kameda, T.; Tang, N. Estrogenic/antiestrogenic activities of polycyclic aromatic hydrocarbons and their monohydroxylated derivatives by yeast two-hybrid assay. *J. Health Sci.* **2007**, *53*, 562-570. DOI: 10.1248/jhs.53.562.


Figure S1. Box plot of seasonal atmospheric concentrations of PAHs in Kanazawa.




The top horizontal lines represent the maximum concentration and bottom horizonal lines represent the minimum concentration. The lines in boxes represent the 25th, 50th (median) and 75th percentile concentrations.

Figure S2. Box plot of seasonal atmospheric concentrations of OH-PAHs in Kanazawa.




The top horizontal lines represent the maximum concentration and bottom horizonal lines represent the minimum concentration. The lines in boxes represent the 25th, 50th (median) and 75th percentile concentrations.

Figure S3. PCA score plot in which the first two PCs for atmospheric PAHs.

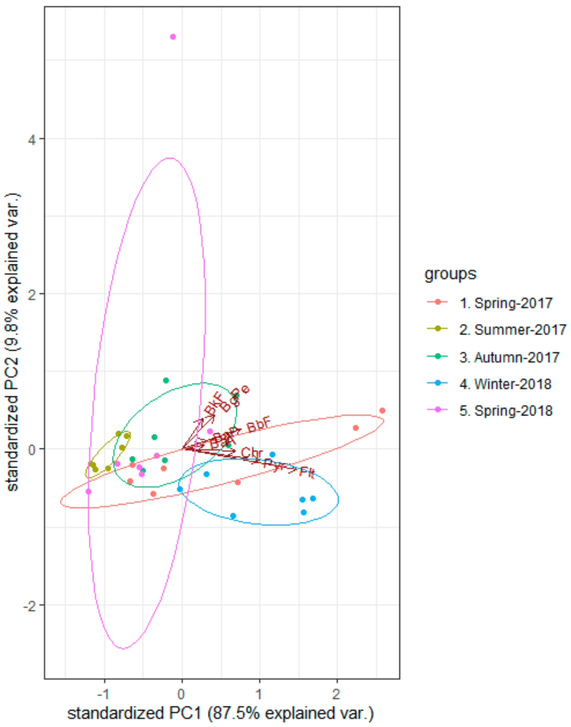


Figure S4. PCA score plot in which the first two PCs for atmospheric OH-PAHs.

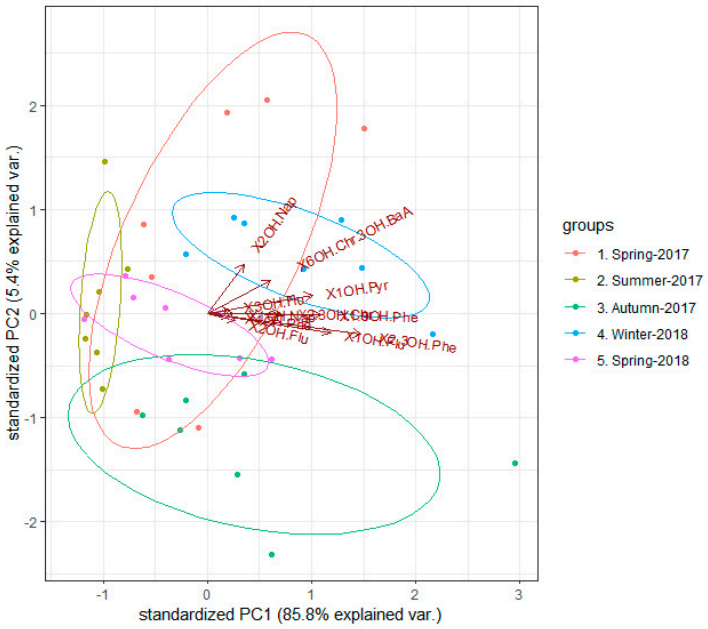



Figure S5. Diagnostic ratios calculated for atmospheric PAHs.

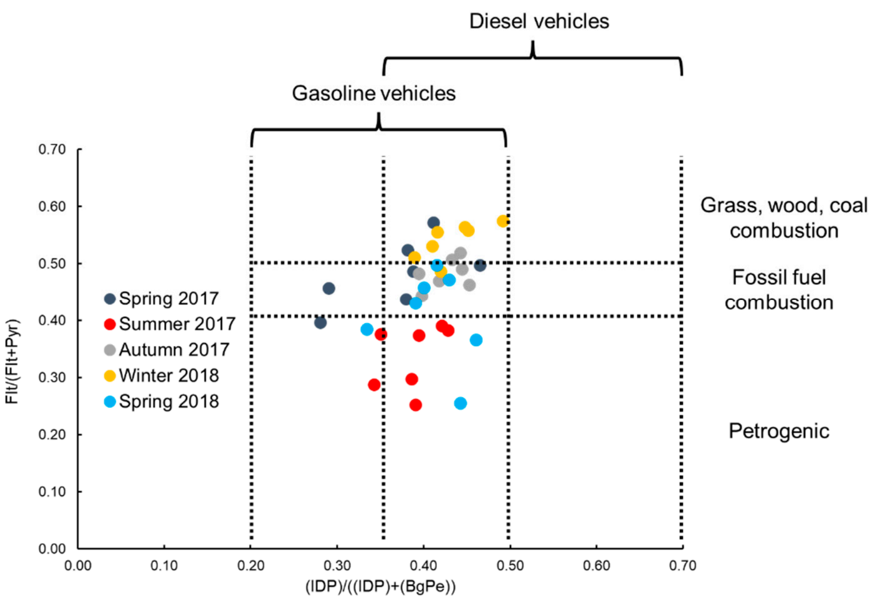


Table S1. Estrogenic and antiestrogenic activities of OH-PAHs from a yeast two-hybrid assay conducted by Hayakawa et al. (2007).

| No. of rings | Abbreviation | $REP_E$ [a] × $10^{-4}$ | $REP_{AE}$ [b] × $10^{-2}$ | No. of rings | Abbreviation | $REP_E$ [a] × $10^{-4}$ | $REP_{AE}$ [b] × $10^{-2}$ |
|---|---|---|---|---|---|---|---|
| 2 | 1OH-Nap | < 1 | < 1 | 5 | 6OH-BbF | < 1 | 3.6 |
| | 2OH-Nap | < 1 | < 1 | | 7OH-BbF | < 1 | < 1 |
| 3 | 2OH-Flu | 5.6 | < 1 | | 8OH-BbF | < 1 | < 1 |
| | 9OH-Flu | < 1 | < 1 | | 9OH-BbF | < 1 | < 1 |
| | 2OH-Phe | 7.5 | < 1 | | 10OH-BbF | < 1 | 7.6 |
| | 3OH-Phe | < 1 | < 1 | | 11OH-BbF | < 1 | < 1 |
| | 4OH-Phe | < 1 | < 1 | | 12OH-BbF | < 1 | < 1 |
| | 9OH-Phe | < 1 | < 1 | | 3OH-BjF | < 1 | < 1 |
| 4 | 3OH-Flt | 4.2 | < 1 | | 3OH-BkF | 1.3 | < 1 |
| | 1OH-Pyr | 2.4 | < 1 | | 8OH-BkF | < 1 | < 1 |
| | 1OH-BaA | < 1 | < 1 | | 9OH-BkF | < 1 | 21 |
| | 2OH-BaA | < 1 | 42 | | 1OH-BaP | < 1 | < 1 |
| | 3OH-BaA | 42 | < 1 | | 2OH-BaP | < 1 | < 1 |
| | 4OH-BaA | 75 | < 1 | | 3OH-BaP | < 1 | < 1 |
| | 5OH-BaA | < 1 | < 1 | | 4OH-BaP | < 1 | < 1 |
| | 9OH-BaA | < 1 | 34 | | 5OH-BaP | < 1 | < 1 |
| | 10OH-BaA | 32 | < 1 | | 6OH-BaP | < 1 | < 1 |
| | 11OH-BaA | < 1 | < 1 | | 7OH-BaP | < 1 | < 1 |
| | 1OH-BcPh | < 1 | < 1 | | 8OH-BaP | < 1 | 43 |
| | 2OH-BcPh | < 1 | 69 | | 9OH-BaP | < 1 | < 1 |
| | 3OH-BcPh | < 1 | 190 | | 10OH-BaP | < 1 | < 1 |
| | 4OH-BcPh | < 1 | 13 | | 11OH-BaP | < 1 | < 1 |
| | 5OH-BcPh | < 1 | 1.2 | | 12OH-BaP | < 1 | < 1 |
| | 1OH-Chr | 4.2 | < 1 | | 4OH-BeP | 4.2 | < 1 |
| | 2OH-Chr | 42 | < 1 | | 9OH-BeP | < 1 | < 1 |
| | 3OH-Chr | < 1 | 40 | | 10OH-BeP | < 1 | < 1 |
| | 4OH-Chr | < 1 | < 1 | | 11OH-BgCh | < 1 | 29 |
| | 6OH-Chr | < 1 | 30 | | 13OH-BgCh | < 1 | 22 |
| 5 | 1OH-BbF | < 1 | < 1 | | 3OH-DB | < 1 | < 1 |
| | 2OH-BbF | < 1 | < 1 | 6 | 6OH-IDP | < 1 | < 1 |
| | 3OH-BbF | < 1 | < 1 | | 8OH-IDP | < 1 | < 1 |

*a*) The estrogenic activity of each test compound was assayed in the concentration range from $1 \times 10^{-6}$ to $1 \times 10^{-9}$ M. The $REP_E$ was calculated as the inverse value of the relative concentration of the test compound that gave the same activity of $E_2$ as a positive control. *b*) The antiestrogenic activity of each test compound was assayed in the concentration range from $1 \times 10^{-6}$ to $1 \times 10^{-9}$ M. The $REP_{AE}$ was calculated as the inverse value of the relative concentration of the test compound that gave the same activity of 4-hydroxytamoxifen as a positive control in the presence of 1 nM $E_2$. Abbreviations: OH-PAHs, hydroxylated polycyclic aromatic hydrocarbons; $REP_E$, relative effective potency of estrogenic activity; $REP_{AE}$, relative effective potency of antiestrogenic activity; Nap, naphthalene; Flu, fluorene; Phe, phenanthrene; Flt, fluoranthene; Pyr, pyrene; BaA, Benz[*a*]anthracene; BcPh, benzo[*c*]phenanthrene; Chr, Chrysene; BbF, Benzo[*b*]fluoranthene; BjF, benzo[*j*]fluoranthene; BkF, Benzo[*k*]fluoranthene; BaP, Benzo[*a*]pyrene; BeP, benzo[*e*]fluoranthene; BgCh, benzo[*g*]chrysene; DB, dibenz[*a,h*]anthracene; and IDP, indeno[*1,2,3-c,d*]pyrene.

Table S2. Binding affinities of PAHs and OH-PAHs to hER conducted by Hayakawa et al. (2007).

| No. of rings | Abbreviation | Relative binding affinity[a)] |
|---|---|---|
| 3 | 2OH-Flu | 0.128 |
| | 2OH-Phe | 0.232 |
| 4 | Flt | 0.005 |
| | 3OH-Flt | 0.207 |
| | Pyr | 0.004 |
| | 1OH-Pyr | 0.110 |
| | BaA | 0.005 |
| | 1OH-BaA | 0.047 |
| | 2OH-BaA | 0.137 |
| | 3OH-BaA | 0.228 |
| | 4OH-BaA | 0.294 |
| | 5OH-BaA | 0.058 |
| | 9OH-BaA | 0.410 |
| | 10OH-BaA | 0.295 |
| | 11OH-BaA | 0.027 |
| | BcPh | 0.002 |
| | 1OH-BcPh | 0.001 |
| | 2OH-BcPh | 0.185 |
| | 3OH-BcPh | 0.345 |
| | 4OH-BcPh | 0.210 |
| | 5OH-BcPh | 0.044 |
| | Chr | 0.003 |
| | 1OH-Chr | 0.126 |
| | 2OH-Chr | 0.406 |
| | 3OH-Chr | 0.138 |
| | 4OH-Chr | 0.012 |
| | 6OH-Chr | 0.090 |
| 5 | 10OH-BbF | 0.115 |
| | 3OH-BkF | 0.150 |
| | 9OH-BkF | 0.205 |
| | 1OH-BaP | 0.187 |
| | 3OH-BaP | 0.394 |
| | 8OH-BaP | 0.376 |
| | 4OH-BeP | 0.117 |
| | 11OH-BgCh | 0.197 |
| | 13OH-BgCh | 0.135 |

*a*) hERa was used in this assay. Relative binding affinity of each test compound ($1 \times 10^{-5}$ M) was calculated from binding affinity of $E_2$ (17β-estradiol, $1 \times 10^{-7}$ M) as a competitor. Abbreviations: PAHs, polycyclic aromatic hydrocarbons; OH-PAHs, hydroxylated PAHs; hER, human estrogen receptor; Flu, fluorene; Phe, phenanthrene; Flt, fluoranthene; Pyr, pyrene; BaA, Benz[*a*]anthracene; BcPh, benzo[*c*]phenanthrene; Chr, Chrysene; BbF, Benzo[*b*]fluoranthene; BkF, Benzo[*k*]fluoranthene; BaP, Benzo[*a*]pyrene; BeP, benzo[*e*]fluoranthene; and BgCh, benzo[*g*]chrysene.