Supplemental

**Long-term and seasonal changes in emission sources of atmospheric particulate-bound pyrene and 1-nitropyrene in selected four cities in the Western Pacific**

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Figure 1: Fig. S1.

Table 2: Table S1, Table S2.



Fig. S1. Sampling cities in Western Pacific Region

Table S1. Characteristics of the sampled cities

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| City | Location | Population(×103) | Avg. temp. (°C) | Characteristics |
| Latitude, Longitude | Summer | Winter |
| Kanazawa(Japan) | 36°33′N, 136°40′E | 450 | 27.0 | 3.8 | Capital of Ishikawa prefecture; Commercial city; Covered with sow in winter |
| Kitakyushu(Japan) | 33°53′N, 130°53′E | 920 | 27.4 | 5.8 | Industrial city including iron manufacturing; Coke oven plants consume coal |
| Shenyang(China) | 41°37′N, 123°25′E | 8,320 | 24.6 | −12.0 | Capital of Liaoning province; Agricultural and industrial city; Coal heating in winter |
| Shanghai(China) | 31°13′N, 116°24′E | 24,760 | 27.8 | 3.5 | Business and commercial city; Economic and financial center |

Text S2. Values of *a* and *b* with atmospheric concentrations of Pyr and 1-NP in four cities from 1997 to 2021

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Year/Season | [Pyr]ng m-3 | [1-NP]pg m-3 | *a* | *b* |
| Kanazawa |
| 1997S | 0.202 | 44.5 | 0.996 | 0.422 |
| 1997W | 0.587 | 119 | 0.995 | 0.391 |
| 1999S | 0.587 | 44.5 | 0.995 | 0.351 |
| 1999W | 0.506 | 119 | 0.996 | 0.450 |
| 2004S | 0.166 | 22.5 | 0.998 | 0.258 |
| 2005W | 0.202 | 24.0 | 0.990 | 0.226 |
| 2007S | 0.107 | 10.4 | 0.986 | 0.181 |
| 2008W | 0.344 | 12.6 | 0.960 | 0.068 |
| 2010S | 0.081 | 3.7 | 0.969 | 0.087 |
| 2010W | 0.303 | 10.6 | 0.958 | 0.065 |
| 2013S | 0.109 | 4.5 | 0.964 | 0.075 |
| 2014W | 0.405 | 6.4 | 0.903 | 0.028 |
| 2017S | 0.080 | 1.1 | 0.917 | 0.033 |
| 2018W | 0.220 | 2.9 | 0.935 | 0.042 |
| 2020W | 0.067 | 1.8 | 0.941 | 0.045 |
| 2020S | 0.017 | 0.84 | 0.970 | 0.091 |
| 2021W | 0.103 | 1.6 | 0.924 | 0.027 |
| Kitakyushu |
| 1997S | 0.303 | 4.0 | 0.646 | 0.017 |
| 1997W | 0.566 | 13.6 | 0.938 | 0.045 |
| 2004S | 0.607 | 5.4 | 0.825 | 0.014 |
| 2005W | 0.607 | 17.3 | 0.963 | 0.079 |
| 2007S | 0.045 | 1.3 | 0.949 | 0.053 |
| 2008W | 0.607 | 7.2 | 0.916 | 0.032 |
| 2010S | 0.178 | 3.2 | 0.916 | 0.032 |
| 2010W | 0.445 | 9.2 | 0.926 | 0.037 |
| 2013S | 0.061 | 0.50 | 0.796 | 0.012 |
| 2014W | 1.76 | 7.7 | 0.641 | 0.005 |
| Shenyang |
| 2001S | 0.607 | 37.1 | 0.976 | 0.115 |
| 2002W | 50.6 | 178 | 0.553 | 0.004 |
| 2007S | 1.52 | 42.0 | 0.946 | 0.051 |
| 2008W | 15.0 | 95.4 | 0.885 | 0.023 |
| 2010S | 0.546 | 23.7 | 0.966 | 0.082 |
| 2010W | 7.49 | 181 | 0.938 | 0.044 |
| 2013S | 1.21 | 22.3 | 0.916 | 0.032 |
| 2014W | 22.3 | 188 | 0.814 | 0.013 |
| 2018A | 3.42 | 26.0 | 0.793 | 0.012 |
| 2019Sp | 2.79 | 10.3 | 0.568 | 0.004 |
| Shanghai |
| 2007S | 0.223 | 371 | 0.994 | 0.318 |
| 2007W | 0.587 | 37.1 | 0.978 | 0.120 |
| 2010S | 0.152 | 8.4 | 0.973 | 0.103 |
| 2010W | 1.13 | 49.4 | 0.967 | 0.082 |
| 2013S | 0.150 | 4.7 | 0.953 | 0.057 |
| 2014W | 2.63 | 42.0 | 0.903 | 0.028 |
| 2015S | 2.20 | 1.9 | 0.819 | 0.014 |
| 2015W | 1.09 | 11.7 | 0.855 | 0.018 |
| 2017S | 0.110 | 4.3 | 0.956 | 0.072 |
| 2018W | 0.850 | 8.9 | 0.851 | 0.017 |

Season: S, summer; W, winter. Sp, spring; A, autumn. Values *a* and *b* were calculated by the NP method.