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*Supplement of*

## **Long-term (2001–2012) trends of carbonaceous aerosols from a remote island in the western North Pacific: an outflow region of Asian pollutants**

**Suresh K. R. Boreddy et al.**

*Correspondence to:* Kimitaka Kawamura ([kkawamura@isc.chubu.ac.jp](mailto:kkawamura@isc.chubu.ac.jp))

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## 1. Statistical analyses

### 1.1. The linear trend equation

The linear trend equation was used to calculate the trend equation of all chemical species and their ratios using linear regression analysis, as

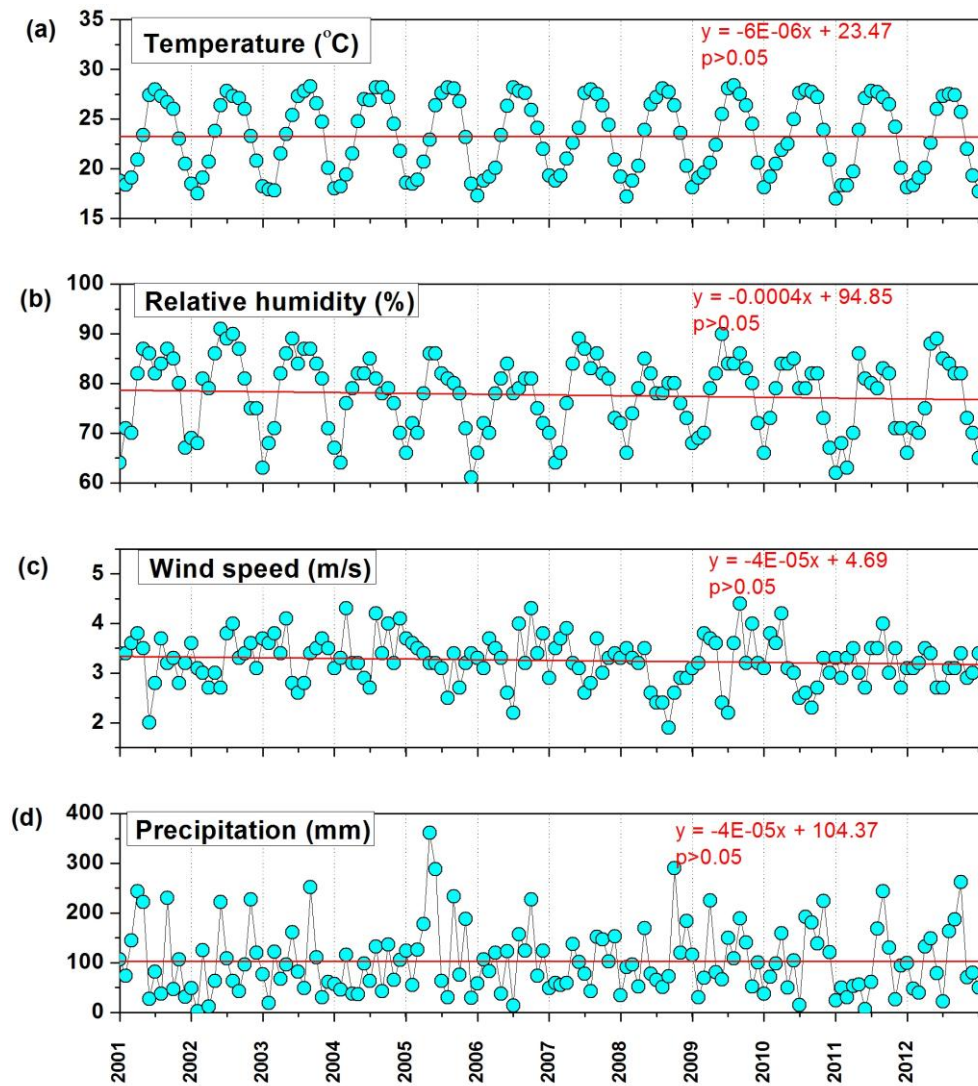
$$y = ax+b \quad (1)$$

where  $y$  is the concentrations in  $\mu\text{g m}^{-3}$ ,  $a$  is the slope,  $x$  is the time in years, and  $b$  is concentrations at the beginning of the period (intercept).

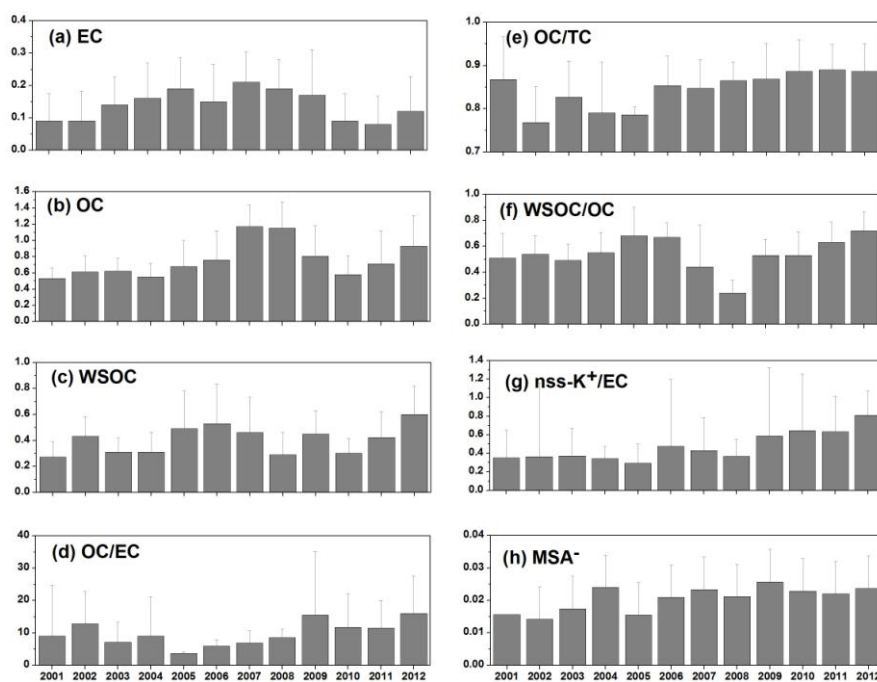
This approach gives results which are simple to interpret; both analytically and graphically on the basis of the shape and parameters of the trend equation. For example, the sign of the concentration trend depends on the value of the slope. In this kind of interpretation when the slope is greater than zero, less than zero, or equal to zero, the sign of the trend is positive (increase), negative (decrease), or there is no trend (no change), respectively.

### 1.2. The Mann-Kendall test

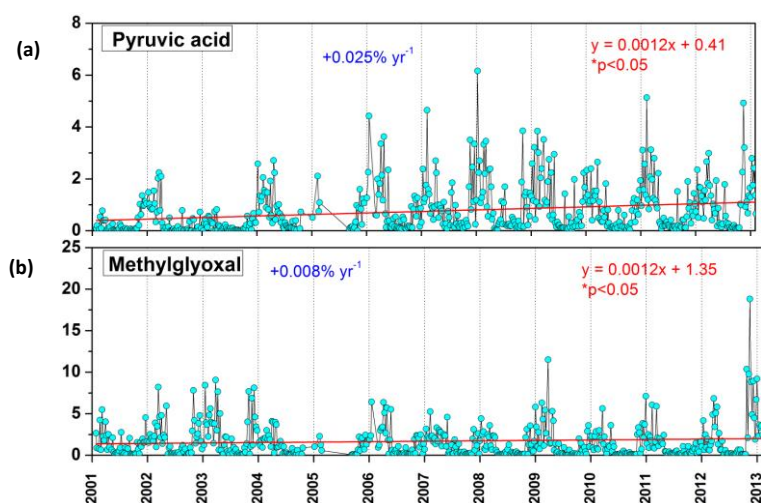
This statistical approach is simple, robust and widely used non-parametric tests to detect the significant trends in time series. According to this approach, two hypotheses were tested: the null hypothesis,  $H_0$ , that there is no trend in the time series; and the alternative hypothesis,  $H_a$ , that there is a significant trend in the series, for a given  $\alpha$  significance level. Probability ( $p$  value) was calculated to determine the level of confidence in the hypothesis. If the  $p$  value is lower than the chosen significance level  $\alpha$  ( $\alpha=5\%$  or  $0.05$ ), the  $H_0$  should be rejected, and  $H_a$  should be accepted (means there is a trend). In case, the  $p$  value is greater than the significance level  $\alpha$ , the  $H_0$  cannot be rejected (there is no trend). In this study, we used XLSTAT software (<http://www.xlstat.com/en/>) for Mann-Kendall test analysis. The absolute value of Kendall  $\tau$  is compared to the standard normal cumulative distribution to define if there is a trend or not at the chosen  $\alpha$  ( $0.05$ ) of significance. A positive and negative value of  $\tau$  indicates an increase and decrease in the trends, respectively.



**Figure S1.** Temporal variations of meteorological parameters such as (a) air temperature (°C), (b) relative humidity (%), (c) wind speed ( $\text{ms}^{-1}$ ), and (d) precipitation (mm) at Chichijima Island during the study period from 2001 to 2012.



**Figure S2.** Annual mean variations ( $\mu\text{g m}^{-3}$ ) of carbonaceous species as well as water-soluble ions and their mass ratios during 2001-2012 over the western North Pacific.



**Figure S3.** Annual trends in the concentrations ( $\text{ng m}^{-3}$ ) of aqueous-phase photooxidations of biogenic isoprene tracer compounds (a) pyruvic acid and (b) methylglyoxal during 2001-2012 over the western North Pacific.