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A sub-decadal trend in diacids in atmospheric aerosols in eastern Asia

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Table S1. Correlations (r^2) of the concentrations of selected major diacids within the studied years (2001 - 2008). The r^2 values were calculated by a linear regression using a least squares fit. The values in the brackets show the p values at the 95% confidence level. The p values were calculated based on the median concentrations using the analysis of variance (ANOVA) technique. The significant trends ($p < 0.05$) have been made bold.

Month	Oxalic acid	Malonic acid	Succinic acid	Adipic acid	Methylsuccinic acid	Phthalic acid
January	0.08 (0.595)	0.02 (0.798)	0.16 (0.440)	0.55 (0.091)	0.17 (0.414)	0.07 (0.613)
February	**0.87 (0.025)**	0.43 (0.159)	0.56 (0.085)	**0.86 (0.008)**	0.38 (0.190)	0.34 (0.200)
March	0.57 (0.143)	0.39 (0.259)	0.74 (0.062)	0.63 (0.104)	0.58 (0.132)	**0.84 (0.027)**
May	0.13 (0.486)	0.05 (0.660)	0.001 (0.990)	0.23 (0.412)	**0.66 (0.048)**	0.55 (0.093)
June	0.11 (0.459)	0.06 (0.588)	0.11 (0.457)	0.09 (0.514)	0.34 (0.172)	0.002 (0.989)
July	0.45 (0.217)	0.04 (0.732)	0.07 (0.662)	0.01 (0.847)	0.01 (0.869)	0.03 (0.779)
August	0.21 (0.443)	0.06 (0.702)	0.07 (0.662)	0.05 (0.712)	0.33 (0.309)	0.19 (0.458)
September	0.03 (0.771)	0.11 (0.583)	0.11 (0.579)	0.11 (0.583)	0.01 (0.917)	0.60 (0.122)
October	0.07 (0.623)	0.04 (0.703)	0.25 (0.313)	0.003 (0.914)	0.20 (0.379)	0.15 (0.450)
November	0.10 (0.492)	0.05 (0.616)	0.20 (0.317)	0.13 (0.419)	0.001 (0.990)	0.04 (0.653)
December	0.26 (0.297)	0.33 (0.230)	0.38 (0.189)	0.43 (0.157)	0.20 (0.370)	**0.69 (0.040)**

Table S2. Correlations (r^2) of the concentrations of a combustion tracer (CO) and precursors (glyoxylic acid, glyoxal and methylglyoxal) of oxalic acid within the studied years (2001 - 2008). The r^2 values were calculated by a linear regression using a least squares fit. The values in the bracket show the p values at the 95% confidence level. The p values were calculated based on the median concentrations using the analysis of variance (ANOVA) technique. The significant ($p < 0.05$) trends have been made bold.

Month	CO	Glyoxylic acid	Glyoxal	Methylglyoxal
January	**0.88 (0.001)**	0.11 (0.512)	0.23 (0.335)	0.54 (0.658)
February	0.04 (0.657)	0.33 (0.236)	0.59 (0.075)	0.28 (0.285)
March	0.24 (0.264)	0.34 (0.305)	0.39 (0.182)	0.004 (0.906)
May	0.15 (0.350)	0.03 (0.746)	**0.70 (0.037)**	0.09 (0.555)
June	0.10 (0.434)	0.27 (0.231)	0.01 (0.888)	0.14 (0.417)
July	0.21 (0.251)	0.03 (0.791)	0.26 (0.302)	0.14 (0.417)
August	0.00 (0.986)	0.40 (0.256)	0.03 (0.743)	0.09 (0.572)
September	**0.50 (0.053)**	0.12 (0.574)	0.24 (0.319)	0.002 (0.904)
October	0.26 (0.242)	0.48 (0.127)	0.09 (0.552)	0.01 (0.881)
November	**0.79 (0.008)**	0.08 (0.536)	0.31 (0.254)	0.44 (0.105)
December	**0.60 (0.040)**	0.36 (0.204)	0.36 (0.206)	0.29 (0.272)

Figure Caption

Fig. 1. Map showing the Gosan site (star symbol) on Jeju Island, South Korea along with the monthly averaged air mass backward trajectories for the time period of 2001-2008. (a) February, (b) March, (c) May, (d) June, e) August, f) September, g) November and h) December. Backward trajectories were calculated using the NOAA HYSPLIT model at 500 m above ground level over 5 days.

Fig. S2. Seasonal variations of malonic (a, b), succinic (c, d), glutaric (e, f) and adipic (g, h) acids in ambient aerosol samples. The right panels are drawn based on the monthly binned concentrations of aerosol samples irrespective of years. The lower and upper whiskers represent the 10th and 90th percentiles, respectively. The outliers are excluded in the right panels to avoid any distortion in the seasonality.

Fig. S3. Seasonal variations of fumaric (a, b), methylmaleic (c, d) and isophthalic (e, f) acids in ambient aerosol samples. The right panels are drawn based on the monthly binned concentrations of aerosol samples irrespective of years. The lower and upper whiskers represent the 10th and 90th percentiles, respectively. The outliers are excluded in the right panels to avoid any distortion in the seasonality.

Fig. S4. The seasonality of total carbon (TC) normalized concentrations of oxalic acid (a, b), azelaic acid (c, d), methylsuccinic acid (e, f) and ketopimelic acid (g, h) in aerosol samples. The right panels are drawn based on the monthly binned normalized-concentrations of aerosol samples irrespective of years. The lower and upper whiskers represent the 10th and 90th percentiles, respectively. The outliers are excluded in the right panels to avoid any distortion in the seasonality.

Fig. S5. The seasonality of total carbon (TC) normalized concentrations of maleic acid (a, b), phthalic acid (c, d) and terephthalic acid (e, f) in aerosol samples. The right panels are drawn based on the monthly binned normalized-concentrations of aerosol samples irrespective of years. The lower and upper whiskers represent the 10th and 90th percentiles, respectively. The outliers are excluded in the right panels to avoid any distortion in the seasonality.

Fig. S6. The seasonality of meteorological parameters over the time period of 2001-2008 at Gosan. (a) precipitation, (b) temperature and (c) relative humidity. The lower whisker represents

the 10th percentile whereas the upper whisker shows the 90th percentile. Data were obtained from Korea Meteorological Administration (KMA).

Fig. S1.

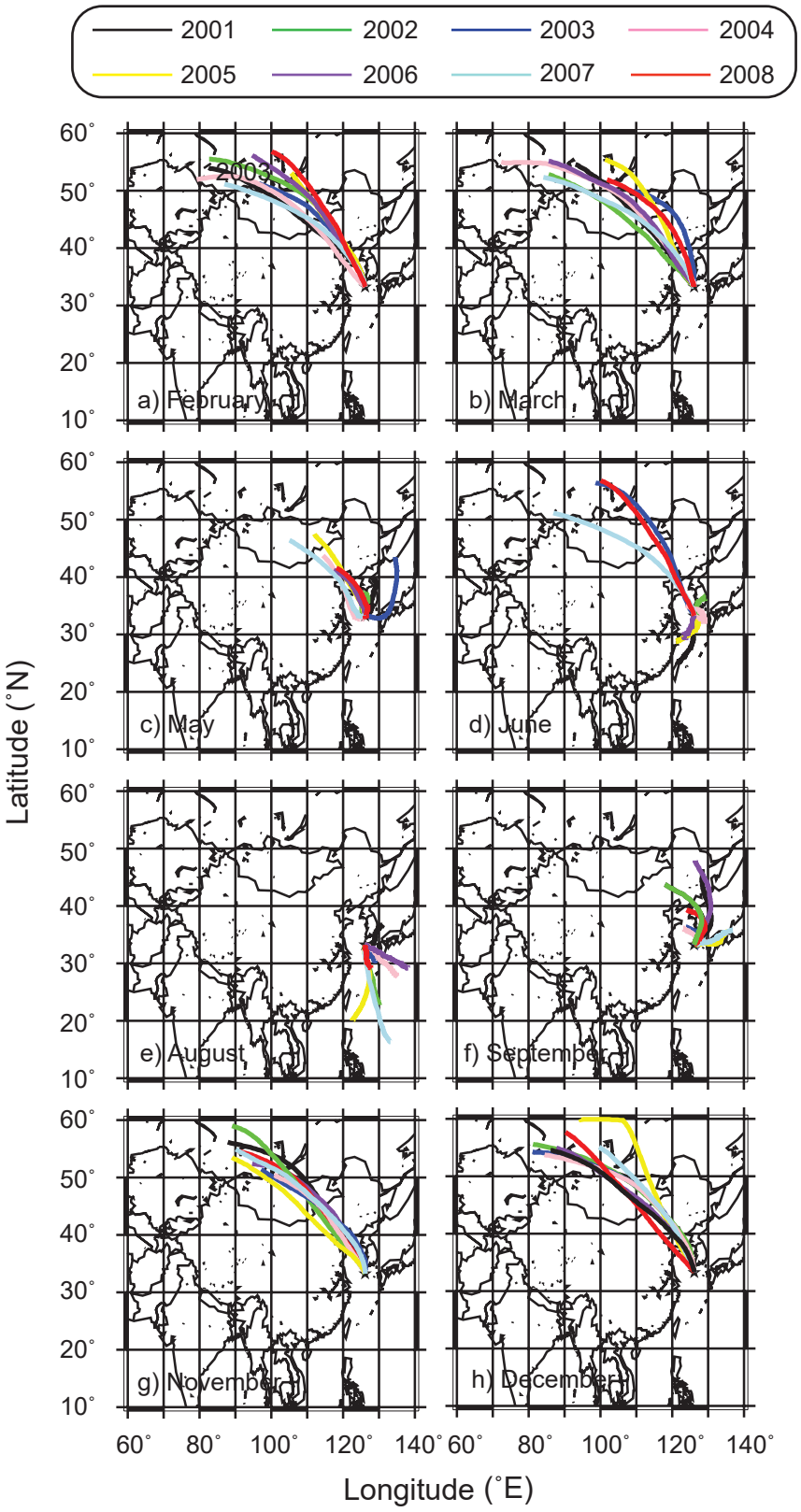

2001
2002
2003
2004
2005
2006
2007
2008
a) February
b) March
c) May
d) June
e) August
f) September
g) November
h) December
Latitude (˚N)
Longitude (˚E)
60˚
50˚
40˚
30˚
20˚
10˚
60˚
80˚
100˚
120˚
140˚

Fig. S2.

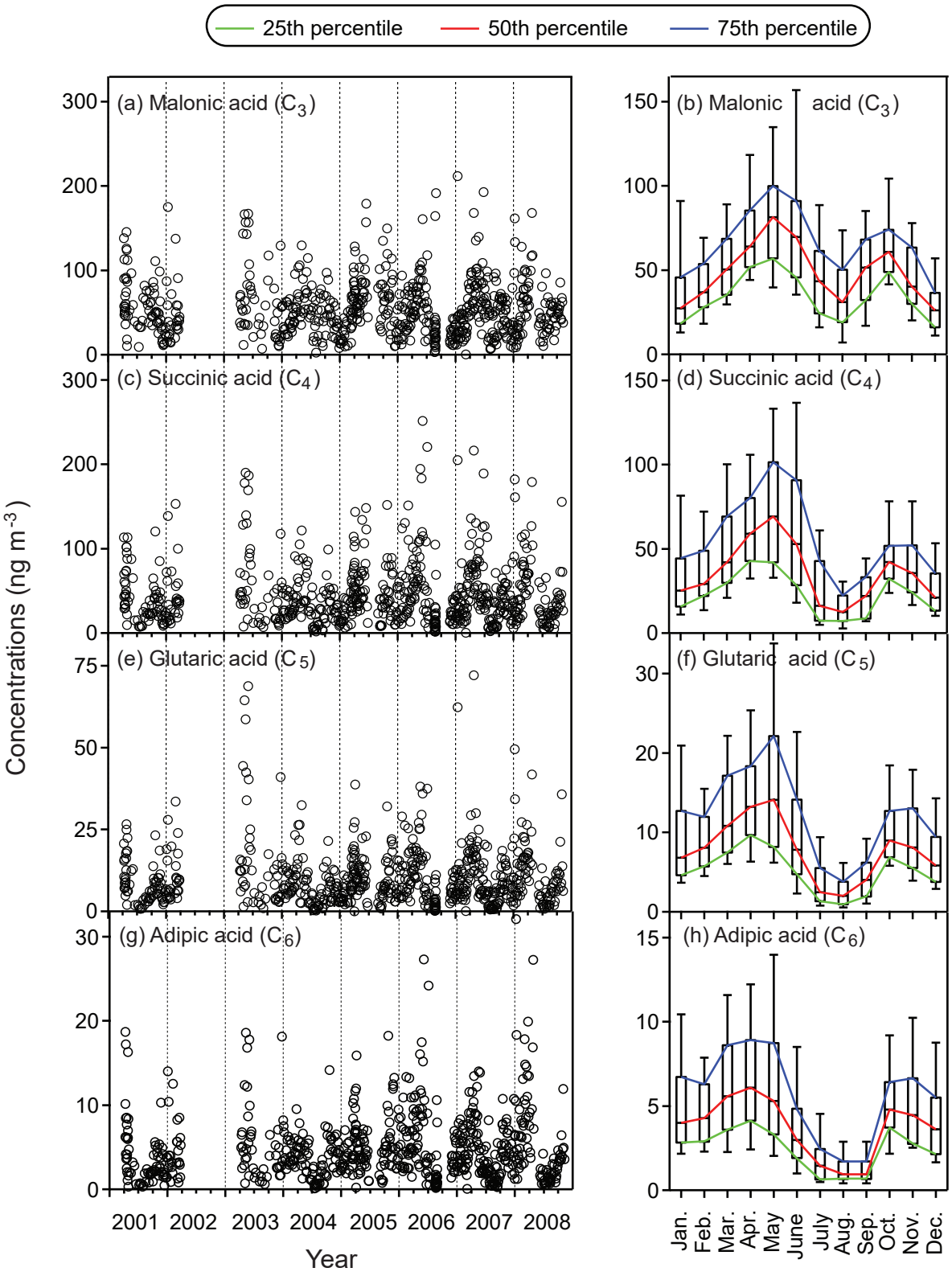

25th percentile
50th percentile
75th percentile
(a) Malonic acid (C_3)
(b) Malonic acid (C_3)
(c) Succinic acid (C_4)
(d) Succinic acid (C_4)
(e) Glutaric acid (C_5)
(f) Glutaric acid (C_5)
(g) Adipic acid (C_6)
(h) Adipic acid (C_6)
Concentrations (ng m^{-3})
2001 2002 2003 2004 2005 2006 2007 2008
Year
Jan. Feb. Mar. Apr. May June July Aug. Sep. Oct. Nov. Dec.

Fig. S3.

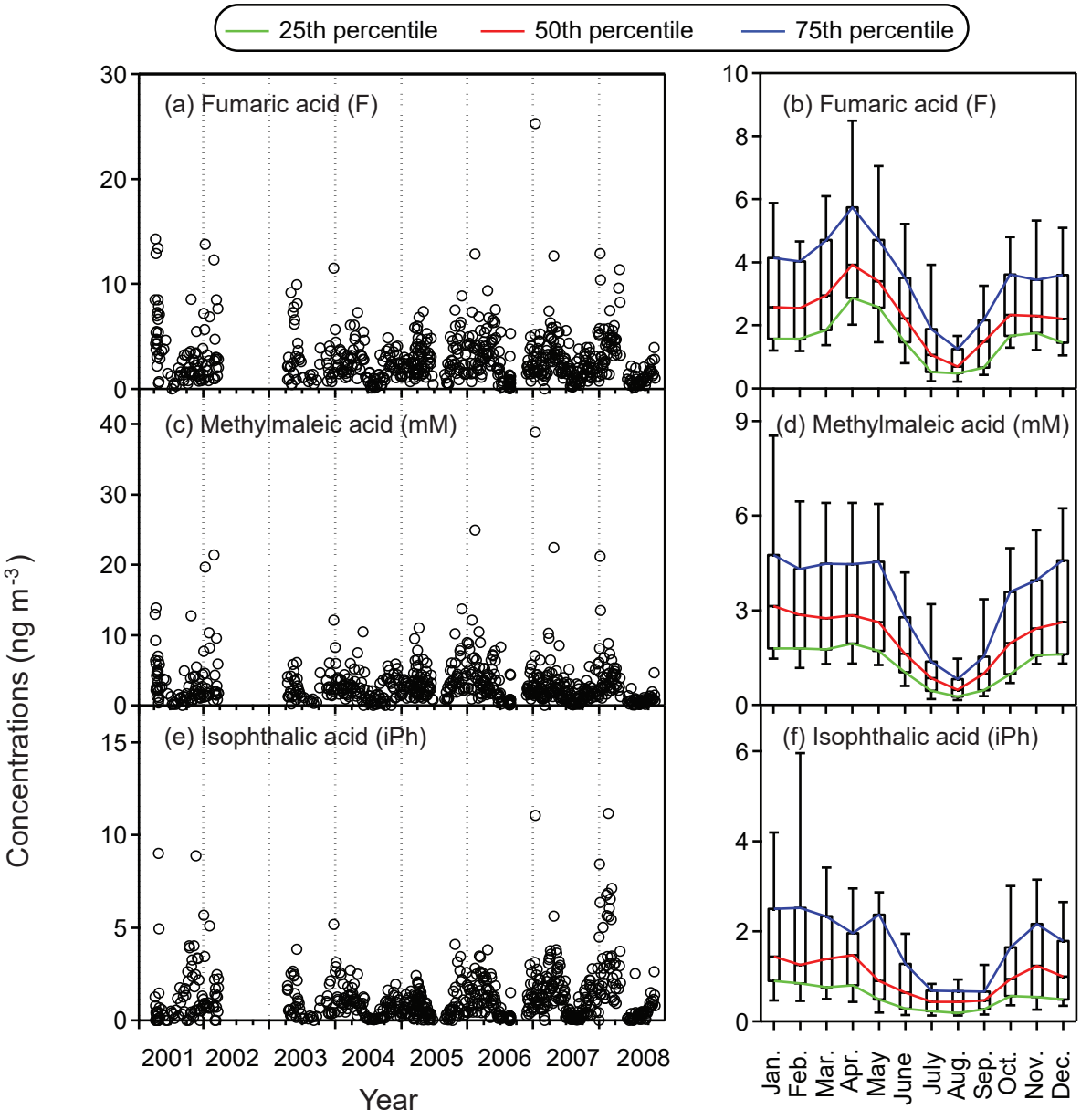

25th percentile
50th percentile
75th percentile
(a) Fumaric acid (F)
(b) Fumaric acid (F)
(c) Methylmaleic acid (mM)
(d) Methylmaleic acid (mM)
(e) Isophthalic acid (iPh)
(f) Isophthalic acid (iPh)
Concentrations (ng m-3)
2001 2002 2003 2004 2005 2006 2007 2008
Year
Jan. Feb. Mar. Apr. May June July Aug. Sep. Oct. Nov. Dec.

Fig. S4.

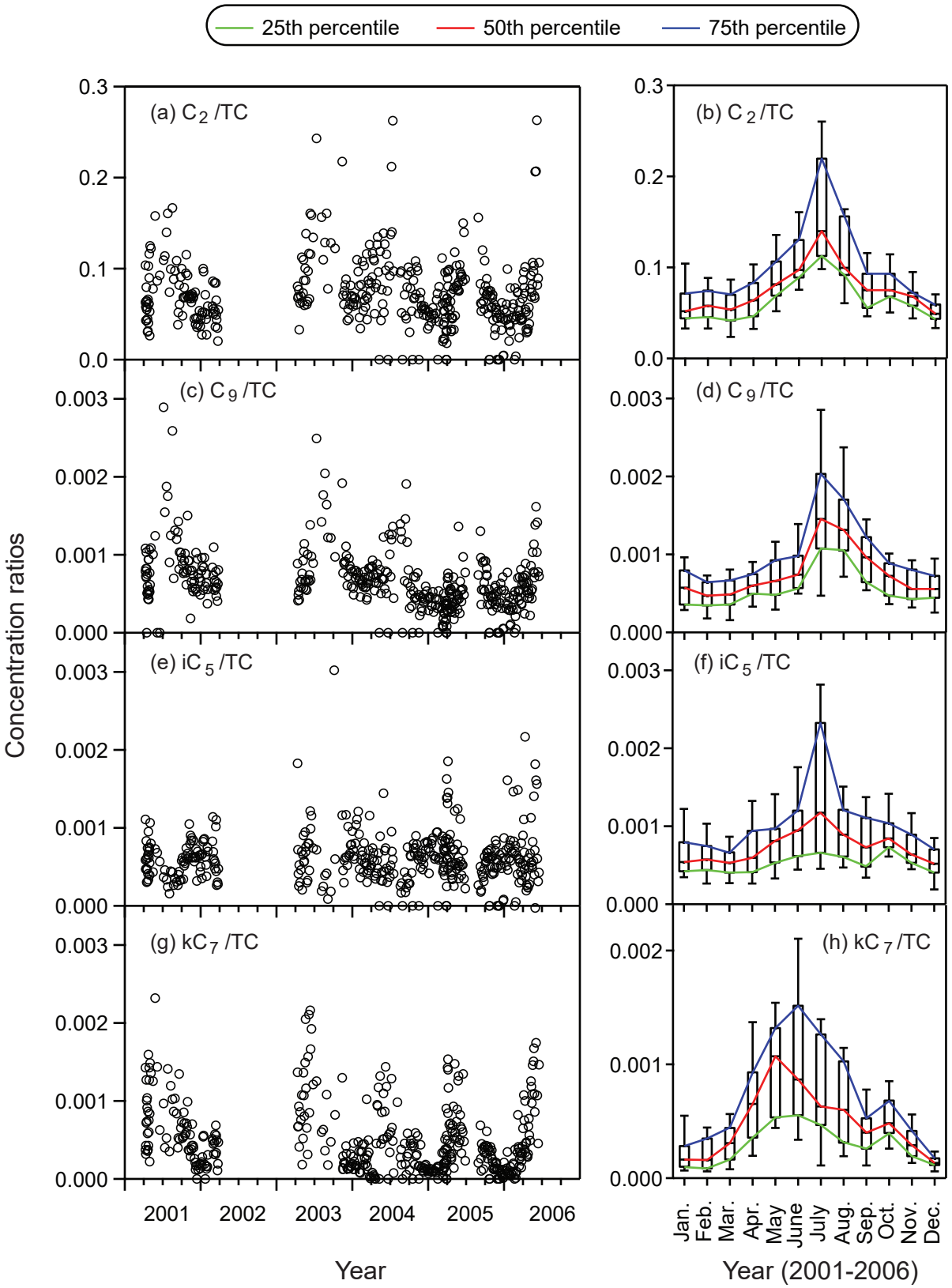

25th percentile
50th percentile
75th percentile
(a) C2 /TC
(b) C2 /TC
(c) C9 /TC
(d) C9 /TC
(e) iC5 /TC
(f) iC5 /TC
(g) kC7 /TC
(h) kC7 /TC
Concentration ratios
Year
Year (2001-2006)
2001
2002
2003
2004
2005
2006
Jan.
Feb.
Mar.
Apr.
May
June
July
Aug.
Sep.
Oct.
Nov.
Dec.

Fig. S5.

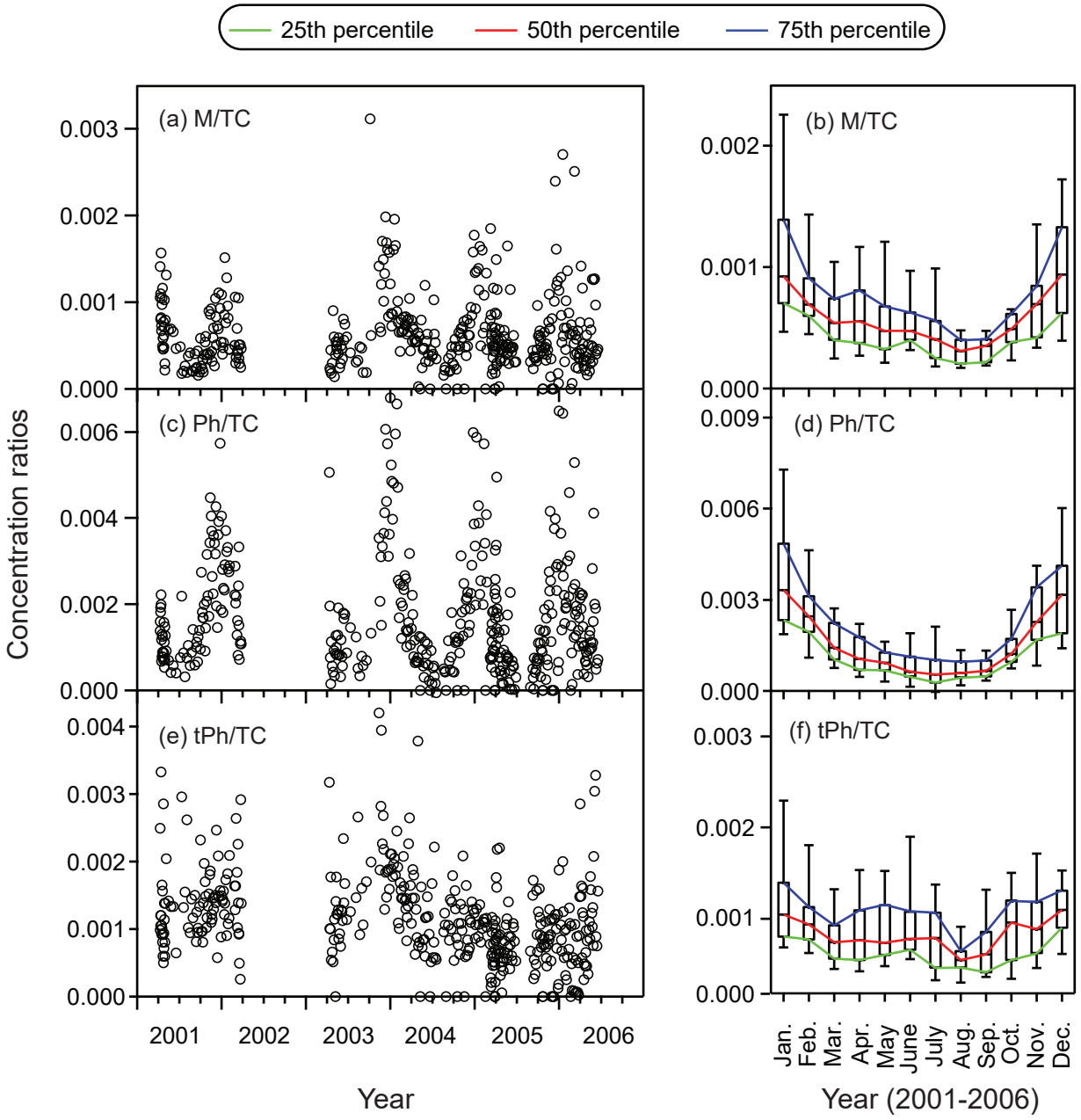
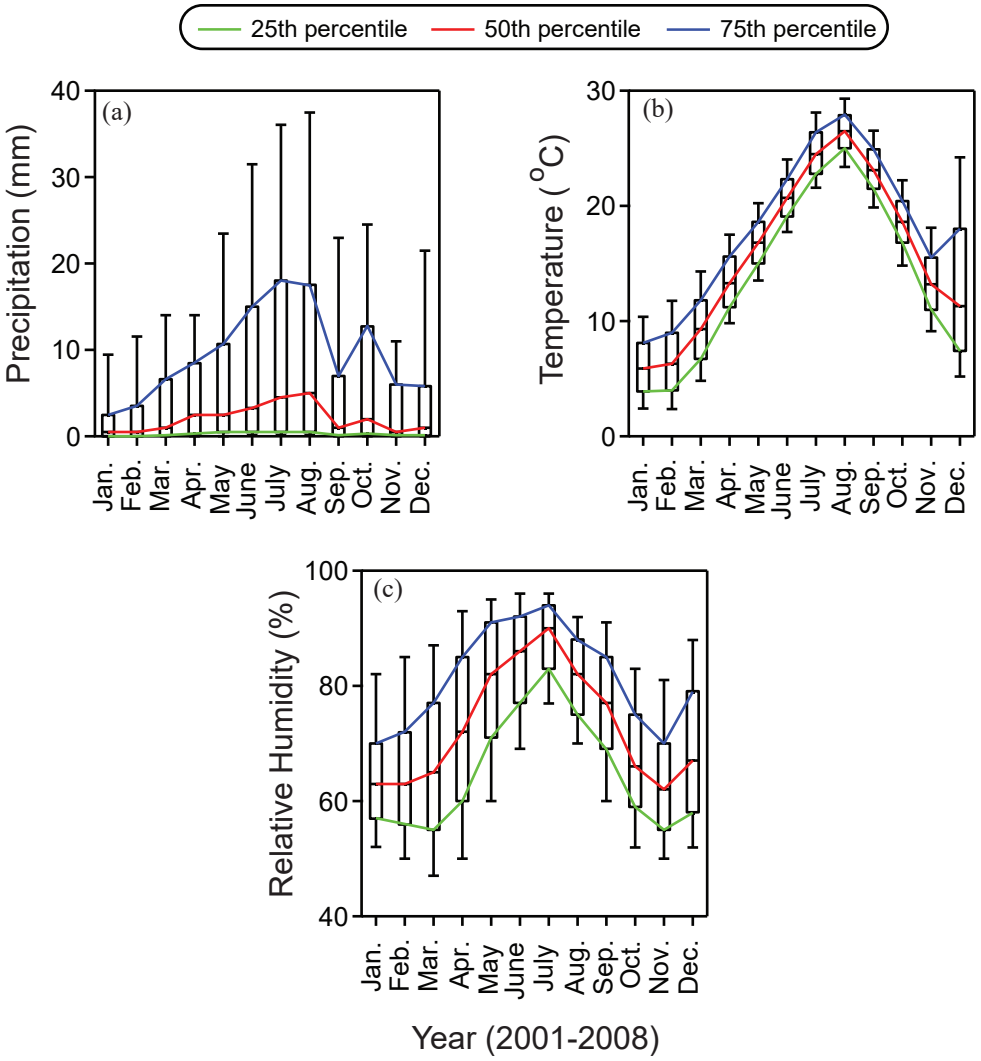

25th percentile
50th percentile
75th percentile
(a) M/TC
(b) M/TC
(c) Ph/TC
(d) Ph/TC
(e) tPh/TC
(f) tPh/TC
Concentration ratios
Year
Year (2001-2006)
2001
2002
2003
2004
2005
2006
Jan.
Feb.
Mar.
Apr.
May
June
July
Aug.
Sep.
Oct.
Nov.
Dec.

Fig. S6.


25th percentile
50th percentile
75th percentile
(a)
Precipitation (mm)
40
30
20
10
0
(b)
Temperature (°C)
30
20
10
0
(c)
Relative Humidity (%)
100
80
60
40
Jan.
Feb.
Mar.
Apr.
May
June
July
Aug.
Sep.
Oct.
Nov.
Dec.
Year (2001-2008)