

## Supplement

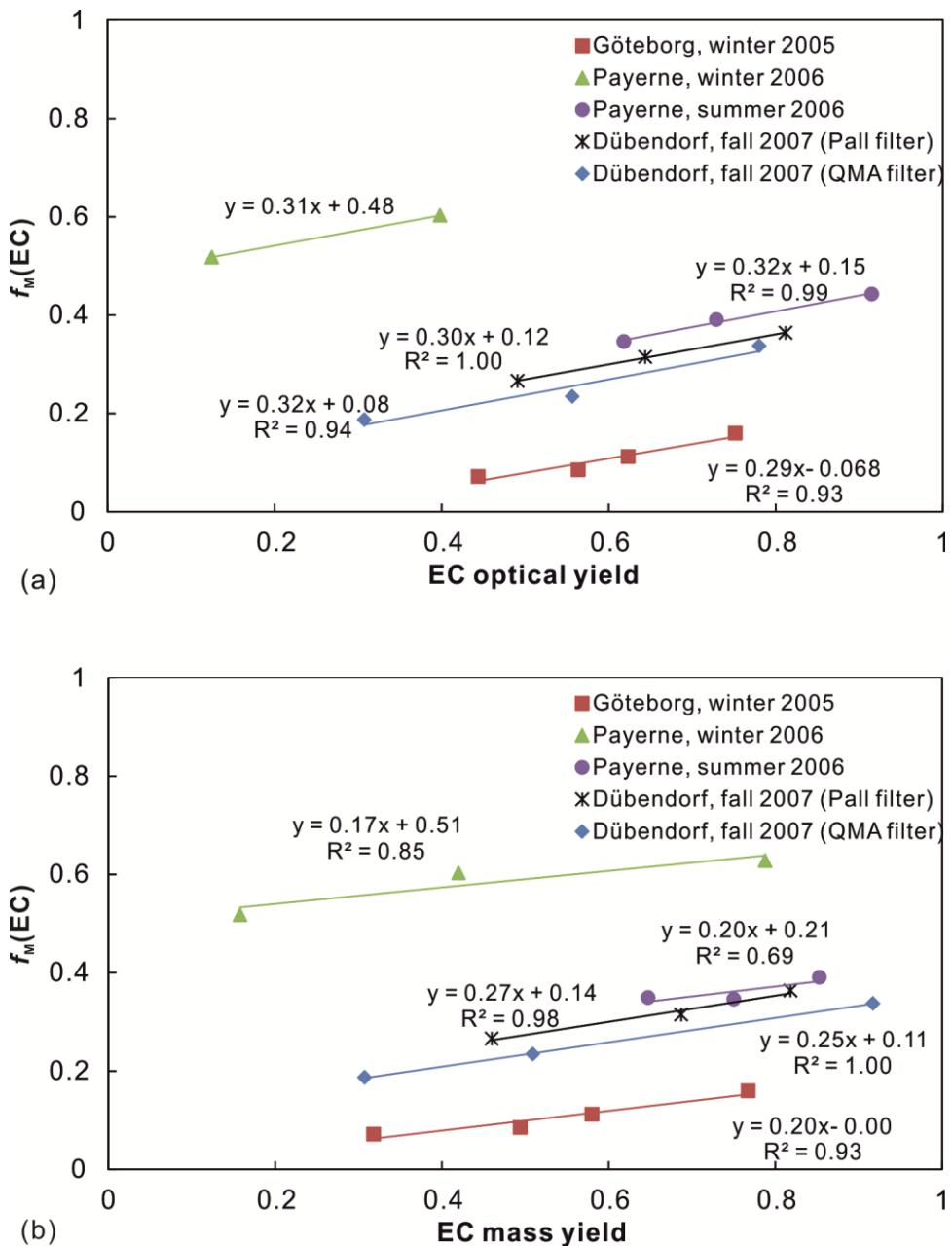


Fig. S1.  $f_m(\text{EC})$  as a function of EC optical yield (a) and EC mass yield (b) for the previous two-step combustion method (THEODORE method) as obtained by analyzing samples with different oven temperatures. For the THEODORE method, the EC optical yield is determined as the ratio of ATN after water extraction and ATN after OC removal in the oven using a white-light aethalometer (Lavanchy et al., 1999). The ratio of rEC (remaining EC after OC removal in the oven) and tEC is regarded as the EC mass yield. Varying slopes between (a) and (b) for identical filters indicate different absorption cross sections for non-rEC and rEC.

**Tab. S1. Estimations of  $f_M$  values of total EC ( $f_M(tEC)$ ) by extrapolation to 100% EC yield based on this work for previously published studies.**

| Site     | Sampling time           | $f_M(rEC)$ | Reference                 | EC yield <sup>a</sup> | $f_M(tEC)$ <sup>b</sup> |
|----------|-------------------------|------------|---------------------------|-----------------------|-------------------------|
| Zurich   | 16-21 Aug 2002          | 0.08±0.03  | Szidat et al., 2004b;     | 0.53±0.08*            | 0.19±0.01               |
| Zurich   | 21-26 Aug 2002          | 0.07±0.02  | Szidat et al., 2006       | 0.59±0.09*            | 0.16±0.01               |
| Zurich   | 19-21 Feb 2003          | 0.25±0.02  |                           | 0.52±0.08*            | 0.36±0.05               |
| Zurich   | 21-23 Feb 2003          | 0.33±0.01  | Szidat et al., 2006       | 0.38±0.06*            | 0.48±0.05               |
| Zurich   | 6-11 Mar 2003           | 0.13±0.01  |                           | 0.27±0.04*            | 0.30±0.06               |
| Zurich   | 21-26 Mar 2003          | 0.16±0.01  |                           | 0.49±0.07*            | 0.27±0.04               |
| Roveredo | 13/14 Jan 2005, Morning | 0.47±0.04  |                           | 0.38±0.06*            | 0.64±0.09               |
| Roveredo | 13/14 Jan 2005, Evening | 0.51±0.02  |                           | 0.68±0.10*            | 0.60±0.06               |
| Roveredo | 19/20 Jan 2005, Morning | 0.53±0.02  |                           | 0.72±0.11*            | 0.60±0.05               |
| Roveredo | 19/20 Jan 2005, Evening | 0.89±0.02  |                           | 0.66±0.10*            | 0.98±0.06               |
| Moleno   | 04/05 Feb 2005, Morning | 0.06±0.02  |                           | 0.54±0.08*            | 0.18±0.07               |
| Moleno   | 04/05 Feb 2005, Evening | 0.22±0.01  | Szidat et al., 2007;      | 0.47±0.07*            | 0.37±0.07               |
| Moleno   | 08/09 Feb 2005, Morning | 0.21±0.01  | Sandradewi et al., 2008a; | 0.64±0.10*            | 0.31±0.06               |
| Moleno   | 08/09 Feb 2005, Evening | 0.17±0.01  | Sandradewi et al., 2008b  | 0.38±0.06*            | 0.34±0.09               |
| Roveredo | 02/03 Mar 2005, Evening | 0.65±0.02  |                           | 0.49±0.07*            | 0.79±0.07               |
| Roveredo | 08/09 Mar 2005, Evening | 0.91±0.05  |                           | 0.68±0.10*            | 0.99±0.07               |
| Roveredo | 09/10 Mar 2005, Morning | 0.24±0.03  |                           | 0.79±0.12*            | 0.29±0.05               |
| Roveredo | 09/10 Mar 2005, Morning | 0.44±0.01  |                           | 0.79±0.12*            | 0.49±0.04               |
| Roveredo | 11/12 Mar 2005, Morning | 0.32±0.02  |                           | 0.67±0.10*            | 0.41±0.06               |
| Roveredo | 4-5 Dec 2005            | 0.86±0.05  |                           | 0.59±0.09*            | 0.97±0.08               |
| Roveredo | 6-7 Dec 2005            | 0.60±0.11  |                           | 0.69±0.10*            | 0.69±0.12               |
| Roveredo | 8-9 Dec 2005            | 0.86±0.08  | Sandradewi et al., 2008a; | 0.64±0.10*            | 0.96±0.09               |
| Roveredo | 11-12 Dec 2005          | 0.76±0.08  | Sandradewi et al., 2008b  | 0.60±0.09*            | 0.87±0.10               |
| Roveredo | 14-15 Dec 2005          | 0.63±0.08  |                           | 0.63±0.10*            | 0.73±0.10               |
| Reiden   | 29-30 Jan 2006          | 0.45±0.03  |                           | 0.58±0.06             | 0.57±0.04               |
| Reiden   | 31 Jan - 01 Feb 2006    | 0.25±0.01  |                           | 0.67±0.07             | 0.35±0.03               |
| Reiden   | 1-2 Feb 2006            | 0.20±0.03  |                           | 0.49±0.05             | 0.36±0.05               |
| Reiden   | 5-6 Feb 2006            | 0.29±0.02  |                           | 0.67±0.07             | 0.40±0.04               |
| Reiden   | 6-7 Feb 2006            | 0.33±0.01  |                           | 0.69±0.07             | 0.43±0.03               |
| Sedel    | 29-30 Jan 2006          | 0.41±0.02  |                           | 0.63±0.06             | 0.52±0.03               |
| Sedel    | 31 Jan - 01 Feb 2006    | 0.29±0.01  | Sandradewi et al., 2008b  | 0.59±0.06             | 0.41±0.03               |
| Sedel    | 1-2 Feb 2006            | 0.24±0.01  |                           | 0.58±0.06             | 0.37±0.03               |
| Sedel    | 5-6 Feb 2006            | 0.54±0.02  |                           | 0.54±0.05             | 0.67±0.04               |
| Sedel    | 6-7 Feb 2006            | 0.39±0.03  |                           | 0.59±0.06             | 0.52±0.05               |
| Zurich   | 12 Jan 2006             | 0.36±0.02  |                           | 0.78±0.08             | 0.42±0.04               |
| Zurich   | 12 Jan 2006             | 0.27±0.02  |                           | 0.50±0.05             | 0.42±0.04               |
| Zurich   | 23-25 Jan 2006          | 0.33±0.02  |                           | 0.81±0.08             | 0.39±0.03               |
| Zurich   | 23-25 Jan 2006          | 0.19±0.03  |                           | 0.47±0.05             | 0.35±0.05               |

**Tab. S1 (continued).**

| <b>Site</b> | <b>Sampling time</b> | $f_M(\text{rEC})$ | <b>Reference</b>    | <b>EC yield<sup>a</sup></b> | $f_M(\text{tEC})^{\text{b}}$ |
|-------------|----------------------|-------------------|---------------------|-----------------------------|------------------------------|
| Göteborg    | 11-14 Feb 2005       | 0.14±0.03         |                     | 0.65±0.06                   | 0.24±0.04                    |
| Göteborg    | 14-18 Feb 2005       | 0.15±0.02         |                     | 0.78±0.08                   | 0.21±0.04                    |
| Göteborg    | 18-25 Feb 2005       | 0.08±0.01         |                     | 0.56±0.06                   | 0.21±0.03                    |
| Göteborg    | 25 Feb - 4 Mar 2005  | 0.11±0.01         |                     | 0.43±0.06*                  | 0.22±0.03                    |
| Råö         | 14-18 Feb 2005       | 0.42±0.06         | Szidat et al., 2009 | 0.63±0.06                   | 0.53±0.07                    |
| Råö         | 18-25 Feb 2005       | 0.35±0.03         |                     | 0.72±0.07                   | 0.43±0.04                    |
| Göteborg    | 13-20 Jun 2006       | 0.13±0.02         |                     | 0.75±0.07                   | 0.20±0.03                    |
| Göteborg    | 20-27 Jun 2006       | 0.17±0.03         |                     | 0.77±0.08                   | 0.24±0.04                    |
| Göteborg    | 27 Jun - 4 Jul 2006  | 0.05±0.02         |                     | 0.79±0.08                   | 0.11±0.03                    |
| Massongex   | 26-27 Nov 2006       | 0.50±0.02         |                     | 0.64±0.06                   | 0.61±0.04                    |
| Massongex   | 27-28 Nov 2006       | 0.36±0.02         |                     | 0.45±0.05                   | 0.52±0.04                    |
| Massongex   | 28-29 Nov 2006       | 0.30±0.01         |                     | 0.53±0.05                   | 0.45±0.04                    |
| Massongex   | 29-30 Nov 2006       | 0.27±0.01         |                     | 0.58±0.06                   | 0.40±0.03                    |
| Massongex   | 1-2 Dec 2006         | 0.28±0.02         |                     | 0.49±0.05                   | 0.44±0.04                    |
| Massongex   | 2-3 Dec 2006         | 0.41±0.01         |                     | 0.79±0.08                   | 0.48±0.03                    |
| Massongex   | 3-4 Dec 2006         | 0.48±0.02         |                     | 0.52±0.05                   | 0.63±0.04                    |
| Massongex   | 5-6 Dec 2006         | 0.29±0.05         |                     | 0.46±0.05                   | 0.46±0.06                    |
| Saxon       | 28-29 Nov 2006       | 0.22±0.01         |                     | 0.48±0.05                   | 0.38±0.04                    |
| Saxon       | 29-30 Nov 2006       | 0.26±0.01         |                     | 0.64±0.06                   | 0.37±0.03                    |
| Saxon       | 1-2 Dec 2006         | 0.28±0.01         |                     | 0.46±0.05                   | 0.44±0.04                    |
| Saxon       | 2-3 Dec 2006         | 0.35±0.01         | Perron et al., 2010 | 0.47±0.05                   | 0.51±0.04                    |
| Saxon       | 3-4 Dec 2006         | 0.48±0.01         |                     | 0.57±0.06                   | 0.61±0.04                    |
| Sion        | 28-29 Nov 2006       | 0.13±0.01         |                     | 0.55±0.05                   | 0.27±0.03                    |
| Sion        | 29-30 Nov 2006       | 0.15±0.01         |                     | 0.58±0.06                   | 0.28±0.03                    |
| Sion        | 1-2 Dec 2006         | 0.19±0.01         |                     | 0.81±0.08                   | 0.25±0.03                    |
| Sion        | 2-3 Dec 2006         | 0.24±0.01         |                     | 0.58±0.06                   | 0.37±0.03                    |
| Sion        | 3-4 Dec 2006         | 0.31±0.02         |                     | 0.39±0.04                   | 0.50±0.05                    |
| Brigerbad   | 28-29 Nov 2006       | 0.18±0.01         |                     | 0.61±0.06                   | 0.30±0.03                    |
| Brigerbad   | 29-30 Nov 2006       | 0.24±0.01         |                     | 0.59±0.06                   | 0.36±0.03                    |
| Brigerbad   | 1-2 Dec 2006         | 0.23±0.01         |                     | 0.48±0.05                   | 0.39±0.04                    |
| Brigerbad   | 2-3 Dec 2006         | 0.37±0.02         |                     | 0.28±0.03                   | 0.59±0.05                    |
| Brigerbad   | 3-4 Dec 2006         | 0.55±0.03         |                     | 0.34±0.03                   | 0.75±0.05                    |
| Mexico City | 21-22 Mar 2006       | 0.15±0.01         |                     | 0.18±0.02                   | 0.25±0.05                    |
| Mexico City | 22-23 Mar 2006       | 0.05±0.02         | Aiken et al., 2010; | 0.37±0.04                   | 0.13±0.04                    |
| Mexico City | 26-27 Mar 2006       | 0.06±0.01         | Hodzic et al., 2010 | 0.29±0.03                   | 0.15±0.04                    |
| Mexico City | 29-30 Mar 2006       | 0.04±0.02         |                     | 0.40±0.04                   | 0.12±0.04                    |

**Tab. S1 (continued).**

| <b>Site</b> | <b>Sampling time</b> | $f_M(\text{rEC})$ | <b>Reference</b>        | <b>EC yield<sup>a</sup></b> | $f_M(\text{tEC})^{\text{b}}$ |
|-------------|----------------------|-------------------|-------------------------|-----------------------------|------------------------------|
| Barcelona   | 27 Feb-1 Mar 2009    | 0.14 $\pm$ 0.01   |                         | 0.95 $\pm$ 0.09             | 0.15 $\pm$ 0.03              |
| Barcelona   | 1-3 Mar 2009         | 0.15 $\pm$ 0.01   |                         | 0.94 $\pm$ 0.09             | 0.17 $\pm$ 0.03              |
| Barcelona   | 13-15 Mar 2009       | 0.14 $\pm$ 0.01   |                         | 0.92 $\pm$ 0.09             | 0.17 $\pm$ 0.03              |
| Barcelona   | 17-19 Mar 2009       | 0.14 $\pm$ 0.01   |                         | 0.91 $\pm$ 0.09             | 0.17 $\pm$ 0.03              |
| Barcelona   | 19-21 Mar 2009       | 0.11 $\pm$ 0.01   |                         | 0.93 $\pm$ 0.09             | 0.13 $\pm$ 0.03              |
| Barcelona   | 21-23 Mar 2009       | 0.24 $\pm$ 0.01   |                         | 0.92 $\pm$ 0.09             | 0.26 $\pm$ 0.03              |
| Barcelona   | 23-25 Mar 2009       | 0.16 $\pm$ 0.01   |                         | 0.89 $\pm$ 0.09             | 0.19 $\pm$ 0.03              |
| Montseny    | 27 Feb-1 Mar 2009    | 0.40 $\pm$ 0.02   |                         | 0.91 $\pm$ 0.09             | 0.43 $\pm$ 0.03              |
| Montseny    | 1-3 Mar 2009         | 0.46 $\pm$ 0.03   |                         | 0.89 $\pm$ 0.09             | 0.50 $\pm$ 0.04              |
| Montseny    | 13-15 Mar 2009       | 0.38 $\pm$ 0.03   | Minguillón et al., 2011 | 0.84 $\pm$ 0.08             | 0.43 $\pm$ 0.04              |
| Montseny    | 17-19 Mar 2009       | 0.32 $\pm$ 0.02   |                         | 0.90 $\pm$ 0.09             | 0.35 $\pm$ 0.03              |
| Montseny    | 19-21 Mar 2009       | 0.25 $\pm$ 0.02   |                         | 0.92 $\pm$ 0.09             | 0.27 $\pm$ 0.03              |
| Montseny    | 21-23 Mar 2009       | 0.47 $\pm$ 0.05   |                         | 0.87 $\pm$ 0.09             | 0.51 $\pm$ 0.05              |
| Montseny    | 23-25 Mar 2009       | 0.34 $\pm$ 0.03   |                         | 0.87 $\pm$ 0.09             | 0.37 $\pm$ 0.04              |
| Barcelona   | 9-10 Jul 2009        | 0.07 $\pm$ 0.02   |                         | 0.92 $\pm$ 0.09             | 0.10 $\pm$ 0.03              |
| Barcelona   | 11-12 Jul 2009       | 0.08 $\pm$ 0.04   |                         | 0.89 $\pm$ 0.09             | 0.11 $\pm$ 0.05              |
| Barcelona   | 16-17 Jul 2009       | 0.10 $\pm$ 0.03   |                         | 0.90 $\pm$ 0.09             | 0.14 $\pm$ 0.04              |
| Barcelona   | 22-23 Jul 2009       | 0.06 $\pm$ 0.02   |                         | 0.88 $\pm$ 0.09             | 0.10 $\pm$ 0.03              |
| Barcelona   | 23-24 Jul 2009       | 0.20 $\pm$ 0.01   |                         | 0.89 $\pm$ 0.09             | 0.23 $\pm$ 0.03              |
| Barcelona   | 28-29 Jul 2009       | 0.05 $\pm$ 0.02   |                         | 0.90 $\pm$ 0.09             | 0.08 $\pm$ 0.04              |
| Montseny    | 9-29 Jul 2009        | 0.23 $\pm$ 0.04   |                         | 0.83 $\pm$ 0.08             | 0.28 $\pm$ 0.05              |

<sup>a</sup> EC yield refers normally to EC optical yield; EC mass yield is used when optical measurements were not available (indicated by an asterisk \*).

<sup>b</sup> In general,  $f_M(\text{tEC})$  is estimated by extrapolation of EC yield to 100% with the individual slope from linear regression of  $f_M(\text{EC})$  and EC yield for the dedicated sampling sites as shown in Figs. 9 and S1. Concerning the samples without the individual slope (i.e. Zurich, Massongex, Saxon, Sion, Barcelona and Montseny), an average slope was used for estimation of  $f_M(\text{tEC})$ .