# Supplementary Material

# Comparison of measurement-based methodologies to apportion secondary organic carbon (SOC) in $PM_{2.5}$: a review of recent studies

Deepchandra Srivastava[1,2,3], Olivier Favez[1], Emilie Perraudin[2,3], Eric Villenave[2,3], Alexandre Albinet[1,*]

[1]INERIS, Parc Technologique Alata, BP 2, 60550 Verneuil-en-Halatte, France; olivier.favez@ineris.fr; deepchandra.srivastava@gmail.com

[2]CNRS, EPOC, UMR 5805 CNRS, 33405 Talence, France

[3]Université de Bordeaux, EPOC, UMR 5805 CNRS, 33405 Talence, France ; eric.villenave@u-bordeaux.fr; emilie.perraudin@u-bordeaux.fr

[*] Correspondence: alexandre.albinet@gmail.com; alexandre.albinet@ineris.fr

**Annex A: Use of EC tracer method**

Calculation of [OC]/[EC] ratio and $[OC]_{non\text{-}comb.}$

The application of the elemental carbon (EC) tracer method requires the measurements of organic carbon ([OC]) and [EC] concentrations together with the determination of the $[OC/EC]_P$ ratio (ratio of OC to EC for the primary sources affecting the site of interest), as well as the non-combustion contribution to the primary OC ($[OC]_{non\text{-}comb}$) [1-3]. $[OC/EC]_P$ and $[OC]_{non\text{-}comb.}$ depend on the dataset used and on the averaging period. The estimation of these parameters is mostly based on linear regressions between [OC] and [EC]. When the studied site is probably not influenced by secondary processes, $[OC/EC]_P$ is directly estimated as the slope of the best fit (linear) and $[OC]_{non\text{-}comb.}$ as the y-intercept. In any other cases, external parameters such as ozone, solar flux, $jNO_2$…are used to determine the most favourable condition for SOA formation or, on the contrary, low solar radiation, low temperature and low $O_3$ concentration levels, and/or occurrence of high NO and low $NO_2$ concentrations highlight the periods of primary emission predominance. In addition, numerical simulations (i.e. OC-EC correlation minimization method) have also been developed to explore the inherent independency between pollutants from primary emissions and secondary sources to derive $[OC/EC]_P$ [4-6].

As observed by Turpin and Huntzicker [2], the linear least-squares is probably not an appropriate method. This linear regression model assumes that the x-values are controlled variables with an exact precision and that all the measurement uncertainties are included in the y variables. However, in the EC tracer method, uncertainties exist for both, [OC] (y variable) and [EC] (x variable). Uncertainties on the x variable result in a significant underestimation of the slope and in a large fictitious positive intercept. Thus, the use of the ordinary least squares is not recommended for the estimation of $[OC/EC]_P$ in the EC tracer method. Deming and York regressions sound more accurate. Both regression models have been formulated to explicitly account for the uncertainties in both coordinates. York regression is the most general while the Deming model is a specific case of the York one [7]. By comparison to the Deming regression, the power of the York regression lies in its ability to use information about measurement uncertainties in the regressed variables to improve the linear fit. Saylor et al. [8] have made corrections on the work done by Chu [9] using the York regression and showed that if data on the measurement uncertainties are available as a function of the measured concentrations, the use of this regression is preferred for the estimation of the parameters of the EC tracer method. If only limited data are available, then the Deming regression should be used. This regression should be also preferred when $[OC]_{non\text{-}comb.}$ is non-null but this tends to overcorrect the problem by slightly overestimating the slope and underestimating the intercept [9]. Recently Wu and Yu [10] also illustrated the drawback of using linear regression techniques those are widely used but often improperly applied due to inappropriate handling of measurement uncertainties. Their work included numerical experiments to evaluate the performance of five linear regression techniques (ordinary least squares, Deming regression, orthogonal distance regression, weighted ordinary least squares and York regression). Results showed good consistency with previous observations, and highlighted that, with appropriate weighting, weighted ordinary least squares, Deming and York regressions provide the best results among all the considered regression analysis methodologies.

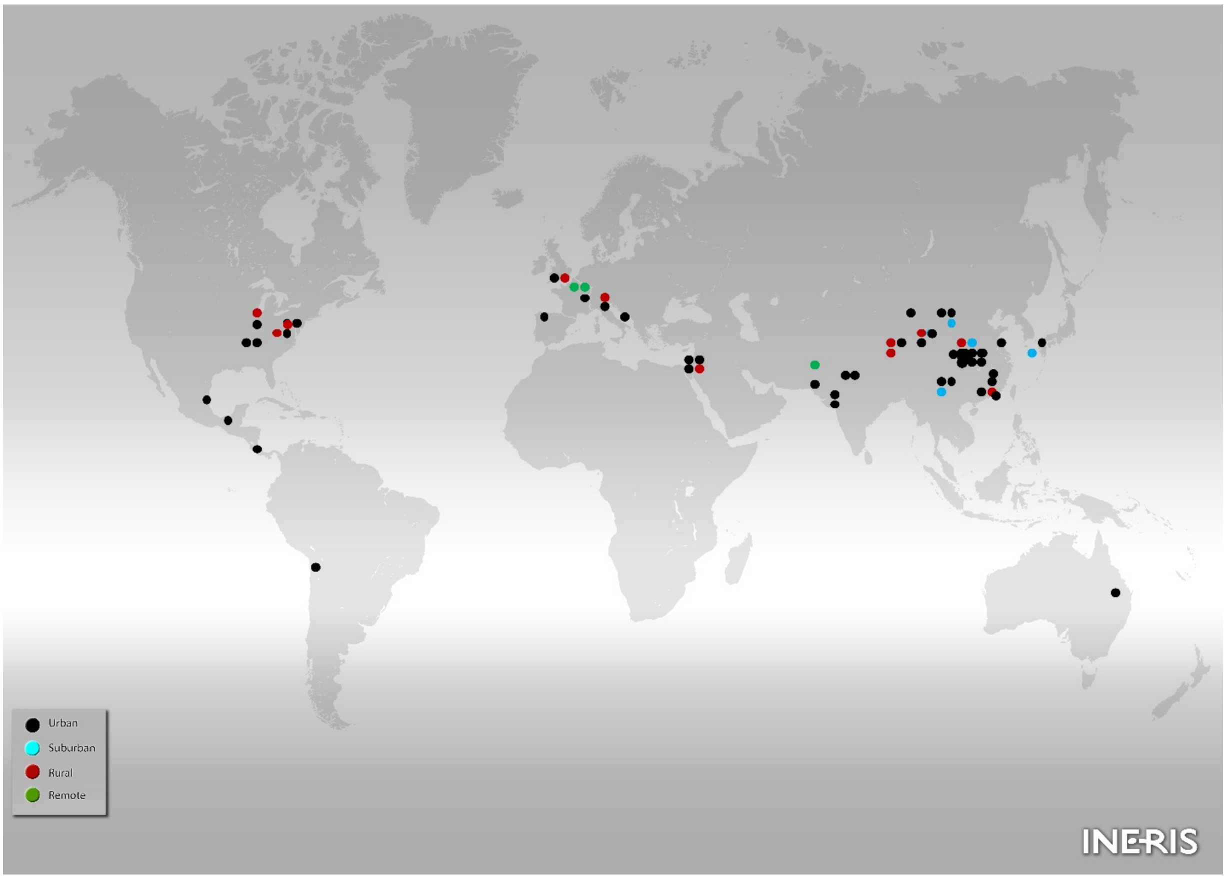


Figure SA1. Location of the monitored sites of the studies considered in this review reporting the use of the elemental carbon (EC) tracer method for the evaluation of secondary organic carbon (SOC) $PM_{2.5}$. In black, urban sites; in blue, suburban sites; in red, rural sites and in green, remote sites.

Table SA1. Reported [OC/EC] ratios in $PM_{2.5}$ for some locations worldwide.

| | Locations | Sampling period | [OC/EC] | References |
|---|---|---|---|---|
| North + South America | Toronto (Canada) | July 2001 | 10.5 | [11] |
| | Vancouver (Canada) | August 2001 | 11.2 | [12] |
| | Seattle (USA) | April - May 1999 | 6.1 | [13] |
| | Pittsburgh (USA) | July 2001 | 4.4 | [14] |
| | Atlanta (USA) | July 2001 | 5.2 | [14] |
| | Chicago (USA) | July 2001 | 3.3 | [14] |
| | New York (USA) | July 2001 | 1.5 | [14] |
| | Monterrey (Mexico) | May - June 2011 | 6.1 | [15] |
| | Monterrey (Mexico) | October - November 2011 | 3.6 | [15] |
| Europe | Helsinki (Finland) | July 2000 - July 2001 | 2.5 | [16] |
| | Barcelona (Spain) | July - December 2004 | 2.6 | [17] |
| | Ghent (Belgium) | June 2005 - February 2005 | 4.1 | [17] |
| | Amsterdam (Netherlands) | July 2005 - February 2006 | 2.9 | [17] |
| | Milan (Italy) | August 2002 - December 2003 | 6.6 | [18] |
| | Budapest (Hungary) | April - May 2002 | 2.1 | [19] |
| | Sonnblick (Austria) | May - June 2003 | 7.9 | [20] |
| | Jungfraujoch (Switzerland) | July - August 1998 | 3.6 | [21] |
| Asia | Mount Abu (India) | March - June 2007 | 3.3 | [22] |
| | Mount Abu (India) | October 2007 - February 2008 | 2.8 | [22] |
| | Pune (India) | April 2012 - March 2013 | 2.4 | [23] |
| | Xiamen (China) | April 2009 | 6.2 | [24] |
| | Xiamen (China) | July 2009 | 4.4 | [24] |
| | Xiamen (China) | October 2009 | 6.6 | [24] |
| | Xiamen (China) | January 2009 | 6.1 | [24] |
| | Beijing (China) | January 2002 - July 2003 | 2.9 | [25] |
| | Beijing (China) | July, November 2002 | 4.6 | [25] |
| | Shanghai (China) | October 2005 - August 2006 | 5 | [26] |
| | Shanghai (China) | October 2005 - August 2006 | 5.6 | [26] |
| | Shanghai (China) | November 2002, August 2003 | 3.8 | [26] |

| | | | |
|---|---|---|---|
| Guangzhou (China) | July - November 2002 | 3.8 | [26] |
| Guangzhou (China) | December 2002, July 2003 | 3.5 | [25] |
| Nanjing (China) | February - September 2001 | 3.6 | [27] |
| Nanjing (China) | February 2001 | 4.9 | [27] |
| Tianjin (China) | Spring 2008 | 3 | [28] |
| Tianjin (China) | Summer 2008 | 1.8 | [28] |
| Tianjin (China) | Fall 2008 | 2.8 | [28] |
| Tianjin (China) | Winter 2008 | 3.8 | [28] |
| Taiyuan (China) | December 2005 - February 2006 | 7 | [29] |
| Hong Kong (China) | August 2004 - March 2005 | 3.5 | [30] |
| Hong Kong (China) | February 2005 - March 2005 | 2.6 | [30] |
| Hong Kong (China) | August - September 2004, February - March 2005 | 5.2 | [30] |
| Mount Heng (China) | March - May 2009 | 5.2 | [31] |
| Mount Tai (China) | March - April 2007 | 5 | [31] |
| Gwangju (Korea) | June - August 2008 | 3.1 | [32] |
| Gwangju (Korea) | December 2008 - February 2009 | 3.0 | [32] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

Table SA2. List of the studies considered for the annual SOC contribution to $PM_{2.5}$ OC estimated using the EC tracer method for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| | Locations | SOC | POC | References |
|---|---|---|---|---|
| North + South America | Atlanta (USA) | 1.5 | 3.3 | [8,14,33,34] |
| | Yorkville (USA) | 1.1 | 2.3 | [8] |
| | Birmingham (USA) | 2.7 | 3.1 | [8,14,34] |
| | Centreville (USA) | 1.1 | 2.9 | [8,34] |
| | Pittsburgh (USA) | 1.4 | 1.5 | [14,35] |
| | Chicago (USA) | 2.3 | 3.3 | [14] |
| | New York City (USA) | 1.1 | 2.4 | [14] |
| | Potsdam (USA) | 1.5 | 0.7 | [36] |
| | Stockton (USA) | 1.6 | 1.2 | [36] |
| | Monterrey (Mexico) | 4.2 | 3.2 | [15] |
| | Costa Rica | 3.9 | 3.5 | [37] |
| | Santiago (Chile) | 3.2 | 7.1 | [38] |
| Europe | Madrid (Spain) | 2.7 | 1.0 | [39,40] |
| | Veneto region (Italy) | 3.8 | 1.7 | [41] |
| | San Pietro Capofiume (Italy) | 2.2 | 2.1 | [42] |
| | Bologna (Italy) | 2.1 | 2.3 | [42] |
| | Thessaloniki (Greece) | 3.7 | 4.4 | [43] |
| | Zloty Potok (Poland) | 3.2 | 5.3 | [44] |
| | Raciborz (Poland) | 4.9 | 8.7 | [44] |
| | Birmingham (UK) | 1.6 | 0.6 | [45,46] |
| | Birmingham (UK) | 1.8 | 0.7 | [45,46] |
| Middle East | Hebron (Palestine) | 2.8 | 2.7 | [47] |
| | Zarqa (Jordan) | 4.9 | 4.2 | [47] |
| | Rahma (Jordan) | 1.4 | 0.8 | [47] |
| | Aqaba (Jordan) | 2.3 | 1.4 | [47] |
| | Amman (Jordan) | 3.4 | 3.3 | [47] |
| | West Jerusalem (Israel) | 2.6 | 1.9 | [47] |
| | Tel Aviv (Israel) | 3.0 | 1.8 | [47] |
| | Haifa (Israel) | 1.8 | 1.7 | [47] |
| | Eilat (Israel) | 2.7 | 0.6 | [47] |
| Asia | Mumbai (India) | 11.4 | 15.9 | [48] |
| | Ahmedabad (India) | 6.1 | 7.3 | [49,50] |
| | Mount Abu (India) | 1.2 | 1.4 | [22] |
| | New Delhi (India) | 26.4 | 34.6 | [51] |
| | Pune (India) | 12.8 | 9.5 | [23,52] |
| | Gurgaon (India) | 13.1 | 15.3 | [53] |
| | Beijing (China) | 5.3 | 8.7 | [54-58] |
| | Shanghai (China) | 4.7 | 6.2 | [26,59-62] |
| | Shanghai (China) | 5.4 | 9.0 | [26,59] |

| | | | |
|---|---|---|---|
| Wangqingsha (China) | 5.4 | 9.8 | [63] |
| Yellow river delta (China) | 3.6 | 4.0 | [64] |
| Nancun (China) | 2.8 | 4.0 | [4] |
| Changchun (China) | 9.6 | 16.4 | [55] |
| Jinchang (China) | 5.8 | 9.9 | [55] |
| Qingdao (China) | 5.8 | 10.0 | [55] |
| Tianjin (China) | 11.7 | 11.9 | [28,55,62,65] |
| Xi'an (China) | 23.9 | 40.9 | [55] |
| Yulin (China) | 8.5 | 14.6 | [55] |
| Chongqing (China) | 11.4 | 21.8 | [55,66] |
| Guangzhou (China) | 5.6 | 8.9 | [30,55,67-69] |
| Hong Kong (China) | 3.1 | 3.0 | [55] |
| Hangzhou (China) | 10.3 | 13.6 | [55] |
| Wuhan (China) | 9.0 | 11.9 | [55] |
| Xiamen (China) | 7.3 | 7.2 | [24,55] |
| Nanjing (China) | 4.9 | 8.9 | [70] |
| Fuzhou (China) | 2.5 | 6.4 | [71,72] |
| Xinhua (China) | 5.9 | 8.2 | [73] |
| Zhaoqing (China) | 3.5 | 4.7 | [67] |
| Chengdu (China) | 5.1 | 13.9 | [66] |
| Neijiang (China) | 4.5 | 13.8 | [66] |
| Haining (China) | 5.6 | 3.4 | [62] |
| Zhongshan (China) | 4.4 | 2.6 | [62] |
| Deyang (China) | 8.6 | 5.2 | [62] |
| Cape Fuguei (Taiwan) | 2.2 | 1.6 | [74] |
| Taipei (Taiwan) | 4.5 | 2.1 | [74] |
| Taichung (Taiwan) | 6.1 | 3.4 | [74] |
| Tainan (Taiwan) | 5.7 | 2.3 | [74] |
| Pingtung (Taiwan) | 7.8 | 2.8 | [74] |
| Penghu (Taiwan) | 1.2 | 1.0 | [74] |
| Hualien (Taiwan) | 2.6 | 1.5 | [74] |
| Chiba Prefecture (Japan) | 1.2 | 2.0 | [75] |
| Yokohama (Japan) | 1.9 | 1.9 | [76] |
| Seoul (Korea) | 1.1 | 6.2 | [77] |
| Gwangju (Korea) | 1.9 | 4.8 | [32] |
| Incheon (Korea) | 4.6 | 3.3 | [78] |
| Brisbane (Australia) | 1.6 | 0.9 | [79] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

Table SA3. List of the studies considered for the spring–summer SOC contribution to $PM_{2.5}$ OC estimated using the EC tracer method for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| | Locations | SOC | POC | References |
|---|---|---|---|---|
| North + South America | Atlanta (USA) | 1.4 | 2.6 | [33,34] |
| | Birmingham (USA) | 1.8 | 2.5 | [34] |
| | Centreville (USA) | 1.2 | 2.8 | [34] |
| | Pittsburgh (USA) | 1.0 | 2.3 | [35] |
| | Santiago (Chile) | 4.7 | 10.1 | [38] |
| | Monterrey (Mexico) | 5.3 | 2.7 | [15] |
| Europe | Milan (Italy) | 4.4 | 0.9 | [18] |
| | Veneto region (Italy) | 0.9 | 1.3 | [41] |
| | Birmingham (UK) | 1.6 | 0.7 | [46] |
| | Birmingham (UK) | 1.6 | 1.0 | [46] |
| | Madrid (Spain) | 2.8 | 0.7 | [39,40] |
| | Raciborz (Poland) | 1.5 | 1.7 | [44] |
| | Zloty Potok (Poland) | 1.7 | 1.2 | [44] |
| | Thessaloniki (Greece) | 2.2 | 3.1 | [43] |
| | Athens (Greece) | 1.6 | 0.5 | [80] |
| | Bologna (Italy) | 1.7 | 1.4 | [42] |
| | San Pietro Capofiume (Italy) | 1.1 | 2.0 | [42] |
| Asia | Ahmedabad (India) | 4.3 | 4.5 | [49,50] |
| | New Delhi (India) | 10.4 | 7.2 | [51] |
| | Pune (India) | 19.4 | 6.4 | [23,52] |
| | Mount Abu (India) | 0.8 | 0.7 | [22] |
| | Guangzhou (China) | 4.0 | 7.3 | [5,30,55,67,68,81] |
| | Beijing (China) | 4.3 | 7.6 | [5,54-58,82,83] |
| | Back Garden (China) | 2.0 | 3.7 | [5] |
| | Shanghai (China) | 2.9 | 6.1 | [26,59-62] |
| | Shanghai (China) | 3.6 | 8.4 | [26,59] |
| | Mount Tai (China) | 11.2 | 5.3 | [84] |
| | Wangqingsha (China) | 3.2 | 4.6 | [63] |
| | Yellow river delta (China) | 4.3 | 3.0 | [64] |
| | Changchun (China) | 5.6 | 6.9 | [55] |
| | Jinchang (China) | 3.6 | 4.5 | [55] |
| | Qingdao (China) | 2.2 | 2.8 | [55] |
| | Tianjin (China) | 6.8 | 7.5 | [28,55,65,85] |
| | Xi'an (China) | 12.2 | 15.1 | [55] |
| | Yulin (China) | 6.1 | 7.6 | [55] |
| | Chongqing (China) | 5.8 | 10.1 | [55,66] |
| | Hong Kong (China) | 2.4 | 1.8 | [55,86] |
| | Hangzhou (China) | 9.1 | 8.0 | [55] |
| | Wuhan (China) | 7.5 | 6.7 | [55] |

| | | | |
|---|---|---|---|
| Xiamen (China) | 5.0 | 4.3 | [24,55] |
| Nanjing (China) | 2.9 | 5.2 | [85] |
| Fuzhou (China) | 2.5 | 6.7 | [71,72] |
| Mount Heng (China) | 1.9 | 1.2 | [31] |
| Zhaoqing (China) | 2.1 | 3.0 | [67] |
| Chengdu (China) | 4.3 | 12.9 | [66] |
| Neijiang (China) | 3.2 | 11.8 | [66] |
| Putian City (China) | 3.1 | 9.9 | [72] |
| Quanzhou City (China) | 2.3 | 7.1 | [72] |
| Shandong (China) | 4.7 | 4.8 | [30] |
| Taishan (China) | 4.7 | 7.9 | [83] |
| Cape Fuguei (Taiwan) | 1.6 | 2.3 | [74] |
| Taipei (Taiwan) | 2.2 | 5.0 | [74] |
| Taichung (Taiwan) | 3.7 | 4.8 | [74] |
| Tainan (Taiwan) | 1.7 | 5.0 | [74] |
| Pingtung (Taiwan) | 2.9 | 6.9 | [74] |
| Penghu (Taiwan) | 1.0 | 1.1 | [74] |
| Hualien (Taiwan) | 1.4 | 2.5 | [74] |
| Chiba Prefecture (Japan) | 1.4 | 1.8 | [75] |
| Yokohama (Japan) | 2.1 | 2.1 | [76] |
| Gwangju (Korea) | 5.7 | 4.6 | [32] |
| Gosan (Korea) | 1.9 | 2.2 | [87] |
| Seoul (Korea) | 1.1 | 5.2 | [77] |
| Incheon (Korea) | 3.6 | 2.9 | [78] |
| Brisbane (Australia) | 1.3 | 1.1 | [79] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

**Annex B. Use of chemical mass balance (CMB).**

Table SB1. List of the molecular markers commonly used to apportion primary aerosol sources using CMB.

| Primary Markers | Primary Sources | References |
|---|---|---|
| Levoglucosan, resin acids, syringaldehyde, acetosyringone, PAHs | Biomass burning | [88-91] |
| Hopanes, PAHs | Gasoline motor vehicles | [88,90,92] |
| Elemental carbon, hopanes, PAHs | Diesel engines | [90,92] |
| C29-C33 n-alkanes with odd carbon preferences | Vegetative detritus | [90,93] |
| n-Hexadecanoic acid, n-octadecanoic acid, 9-hexadecanoic acid, 9-octadecanoic acid, cholesterol | Cooking emissions | [94-96] |
| Syringaldehyde, acetosyringone, syringic acid | Hard wood combustion | [97,98] |
| Resin acids (predominantly dehydroabietic acid, and 7-oxodehydroabietic acid) | Soft wood combustion | [91,97,98] |
| Picene | Coal combustion | [94,99] |
| n-Hentriacontane (C31), n-Dotriacontane (C32), n-Tritriacontane (C33) | Cigarette smoke | [94] |


Figure SB1. Location of the monitored sites of the studies considered in this review reporting the use of the CMB approach for the evaluation of SOC in $PM_{2.5}$. In black, urban sites; in blue, suburban sites; in red, rural sites and in green, remote sites.

Table SB2. List of the studies considered for the annual SOC contribution to $PM_{2.5}$ OC estimated using CMB for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| | Locations | SOC | POC | References |
|---|---|---|---|---|
| North + South America | Santiago (Chile) | 2.7 | 6.4 | [100] |
| | Central LA (USA) | 1.2 | 2.1 | [101-103] |
| | Anaheim (USA) | 0.2 | 2.6 | [103] |
| | Riverside (USA) | 2.2 | 1.1 | [101] |
| | Texas (USA) | 1.6 | 0.9 | [104] |
| | Texas (USA) | 1.1 | 0.7 | [104] |
| | Pensacola (USA) | 4.1 | 2.9 | [105] |
| | Atlanta (USA) | 2.7 | 2.4 | [33,105,106] |
| | Birmingham (USA) | 4.6 | 5.2 | [105] |
| | Charlotte (USA) | 1.6 | 2.2 | [89] |
| | Winston- Salem (USA) | 1.3 | 2.3 | [89] |
| | Hickory (USA) | 1.1 | 3.0 | [89] |
| | Lexington (USA) | 1.3 | 2.5 | [89] |
| | Centreville (USA) | 4.1 | 2.2 | [105] |
| | Northern Minnesota (USA) | 0.0 | 0.8 | [107] |
| | Southern Minnesota (USA) | 0.0 | 1.1 | [107] |
| | Mille Lacs Lake (USA) | 0.5 | 1.2 | [107] |
| | Minneapolis (USA) | 0.1 | 2.9 | [107] |
| | St. Paul (USA) | 0.1 | 2.7 | [107] |
| | Rochester (USA) | 0.3 | 1.8 | [107] |
| Europe | London (UK) | 1.2 | 2.2 | [108] |
| | Birmingham (UK) | 1.2 | 1.6 | [108,109] |
| | Birmingham (UK) | 0.9 | 1.6 | [108,109] |
| | Milan (Italy) | 0.7 | 6.0 | [110] |
| Middle East | Nablus (Palestine) | 3.8 | 4.8 | [111] |
| | Hebron (Palestine) | 3.0 | 2.8 | [111] |
| | East Jerusalem (Palestine) | 1.9 | 3.5 | [111] |
| | Zarqa (Jordan) | 4.9 | 4.2 | [111] |
| | Rahma (Jordan) | 1.4 | 0.8 | [111] |
| | Aqaba (Jordan) | 2.3 | 1.4 | [111] |
| | Amman (Jordan) | 3.4 | 3.3 | [111] |
| | West Jerusalem (Israel) | 2.0 | 2.7 | [111] |
| | Tel Aviv (Israel) | 3.0 | 1.8 | [111] |
| | Haifa (Israel) | 1.8 | 1.7 | [111] |
| | Eilat (Israel) | 2.2 | 1.1 | [111] |
| | Baghdad (Iraq) | 5.5 | 7.5 | [112] |
| Asia | Bishkek (Kyrgyzstan) | 0.7 | 0.8 | [113] |
| | Teploklyuchenka (Kyrgyzstan) | 1.0 | 0.5 | [113] |
| | Lahore (Pakistan) | 7.7 | 53.6 | [114] |

| | | | |
|---|---|---|---|
| Kanpur (India) | 4.8 | 4.5 | [115] |
| Agra (India) | 3.3 | 6.9 | [115] |
| Godavari (Nepal) | 3.4 | 1.7 | [116] |
| Harbin (China) | 2.1 | 6.3 | [117] |
| Tianjin (China) | 1.2 | 7.2 | [118] |
| Hong Kong (China) | 1.6 | 1.7 | [119] |
| Hong Kong (China) | 2.3 | 3.6 | [119,120] |
| Guangzhou (China) | 3.1 | 10.9 | [121] |
| Changhua (China) | 3.3 | 6.1 | [120] |
| Zhongshan (China) | 2.1 | 8.5 | [120] |
| Shenzhen (China) | 2.8 | 8.0 | [120] |
| Gwangju (Korea) | 1.9 | 4.6 | [122] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

Table SB3. List of the studies considered for the spring-summer SOC contribution to $PM_{2.5}$ OC estimated using CMB for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| | **Locations** | **SOC** | **POC** | **References** |
|---|---|---|---|---|
| North + South America | Central LA (USA) | 0.7 | 2.3 | [102,103] |
| | Anaheim (USA) | 0.2 | 1.9 | [103] |
| | Atlanta (USA) | 3.2 | 0.8 | [106,123] |
| | Tennessee Valley (USA) | 2.8 | 1.1 | [124] |
| | Yorkville (USA) | 2.4 | 0.4 | [123] |
| | Pittsburgh (USA) | 2.7 | 0.9 | [125] |
| | Mexico City (Mexico) | 3.3 | 5.3 | [116] |
| | Santiago (Chile) | 2.5 | 8.0 | [100] |
| | Summit (Greenland) | 0.1 | 0.0 | [126] |
| Europe | Birmingham (UK) | 1.5 | 1.6 | [108] |
| | Birmingham (UK) | 1.2 | 1.7 | [108] |
| | Marseille (France) | 3.7 | 1.0 | [127] |
| | Milan (Italy) | 1.4 | 2.7 | [110,128] |
| | Oasi Le Bine (Italy) | 1.1 | 1.4 | [128] |
| | Alps (Italy) | 1.2 | 2.8 | [128] |
| Middle East | West Jerusalem (Israel) | 1.9 | 2.9 | [129] |
| | East Jerusalem (Israel) | 2.5 | 2.7 | [129] |
| Asia | Bishkek (Kyrgyzstan) | 1.5 | 0.4 | [113] |
| | Teploklyuchenka (Kyrgyzstan) | 0.8 | 0.3 | [113] |
| | Lahore (Pakistan) | 3.7 | 29.1 | [114] |
| | Kanpur (India) | 5.4 | 8.1 | [115] |
| | Agra (India) | 4.3 | 5.9 | [115] |
| | Godavari (Nepal) | 3.1 | 1.6 | [116] |
| | Harbin (China) | 1.1 | 2.8 | [117] |
| | Hong Kong (China) | 1.2 | 1.1 | [119,120] |
| | Hong Kong (China) | 2.9 | 2.4 | [119,120] |
| | Guangzhou (China) | 4.6 | 10.8 | [120] |
| | Changhua (China) | 4.8 | 5.6 | [120] |
| | Zhongshan (China) | 3.7 | 4.1 | [120] |
| | Shenzhen (China) | 3.8 | 5.6 | [120] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

**Annex C. Use of the secondary organic aerosol (SOA) tracer method**

Table SC1. Details of the SOA tracers considered in the SOA tracer method and laboratory generated aerosol mass fractions [130].

| Organic markers | Precursors | MW (g mol$^{-1}$) | $f_{SOA}$ | $f_{SOC}$ | SOA/SOC |
|---|---|---|---|---|---|
| 2-Methylglyceric acid | Isoprene | 134 | 0.063±0.016 | 0.155±0.039 | 1.37±0.15 |
| 2-Methylthreitol | | 136 | | | |
| 2-Methylerythritol | | 136 | | | |
| 3-Isopropylpentanedioic acid | $\alpha$-Pinene | 174 | 0.168±0.081 | 0.231±0.111 | 1.98±0.14 |
| 3-Acetylpentanedioic acid | | 174 | | | |
| 2-Hydroxy-4-isopropyladipic acid | | 204 | | | |
| 3-Acetylhexanedioic acid | | 188 | | | |
| 3-Hydroxyglutaric acid | | 148 | | | |
| 2-Hydroxy-4,4-dimethylglutaric acid | | 176 | | | |
| 3-(2-Hydroxy-ethyl)-2,2-dimethylcyclobutane-carboxylic acid | | 172 | | | |
| Pinic acid | | 186 | | | |
| Pinonic acid | | 184 | | | |
| 2,3-Dihydroxy-4-oxopentanoic acid (DHOPA) | Toluene | 148 | 0.040±0.0013 | 0.079±0.0026 | 2.47±0.55 |
| β-Caryophyllinic acid | β-Caryophyllene | 254 | 0.0109±0.0022 | 0.0230±0.0046 | 2.11±0.65 |

Table SC2. List of the studies considered for the annual SOC contribution to $PM_{2.5}$ OC estimated using the SOA tracer method for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| **Locations** | **$SOC_{isoprene}$** | **$SOC_{\alpha\text{-pinene}}$** | **$SOC_{toluene}$** | **$SOC_{\beta\text{-caryophyllene}}$** | **POC** | **References** |
|---|---|---|---|---|---|---|
| Research Triangle Park (USA) | 0.5 | 0.3 | 0.2 | 0.2 | 2.5 | [130,131] |
| Cincinnati (USA) | 0.6 | 0.1 | 0.2 | 0.2 | 1.7 | [132] |
| Detroit (USA) | 0.3 | 0.3 | 0.3 | 0.2 | 2.8 | [132] |
| Bondville (USA) | 0.3 | 0.1 | 0.2 | 0.2 | 0.7 | [132] |
| East St. Louis (USA) | 0.9 | 0.1 | 0.2 | 0.2 | 2.9 | [132] |
| Northbrook (USA) | 0.2 | 0.2 | 0.2 | 0.1 | 1.7 | [132] |
| Nam Co Lake (Tibet) | 0.1 | 0.02 | 0.1 | 0.01 | 1.5 | [133] |
| Shanghai (China) | 0.1 | 0.02 | 0.2 | 0.03 | 10.2 | [59] |
| Shanghai (China) | 0.1 | 0.02 | 0.2 | 0.1 | 11.1 | [59] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

Table SC3. List of the studies considered for the spring-summer SOC contribution to $PM_{2.5}$ OC estimated using the SOA tracer method for all the monitored sites from 2006 to 2016. SOC and POC concentrations in μgC $m^{-3}$.

| Locations | | $SOC_{isoprene}$ | $SOC_{\alpha\text{-pinene}}$ | $SOC_{toluene}$ | $SOC_{\beta\text{-caryophyllene}}$ | POC | References |
|---|---|---|---|---|---|---|---|
| North + South America | Research Triangle park (USA) | 0.9 | 0.7 | 0.4 | 0.9 | 1.6 | [130,134] |
| | Cincinnati (USA) | 1.1 | 0.2 | 0.3 | 0.2 | 1.5 | [132,134] |
| | Detroit (USA) | 0.5 | 0.3 | 0.6 | 0.2 | 2.0 | [90,132,134] |
| | Cleveland (USA) | 0.5 | 0.2 | 0.6 | 0.1 | 1.4 | [90,132,134] |
| | Medina (USA) | 0.8 | 0.3 | 0.3 | 0.1 | 0.01 | [134] |
| | Bondville (USA) | 0.7 | 0.2 | 0.3 | 0.2 | 0.4 | [132,134] |
| | East St. Louis (USA) | 0.8 | 0.2 | 0.2 | 0.1 | 2.6 | [132,134] |
| | Northbrook (USA) | 0.4 | 0.2 | 0.3 | 0.2 | 1.0 | [132,134] |
| | Bakersfield (USA) | 0.1 | 0.1 | 0.1 | - | 5.0 | [134] |
| | Pasadena (USA) | 0.04 | 0.1 | 0.1 | - | 3.2 | [134] |
| | Riverside (USA) | 0.1 | 0.2 | 0.9 | 0.1 | 4.2 | [34,90] |
| | Mexico City (Mexico) | 0.2 | 0.2 | 1.9 | 0.2 | 3.5 | [88] |
| | Pensacola (USA) | 0.7 | 0.5 | 0.3 | 0.04 | 4.3 | [34,134] |
| | Centreville (USA) | 1.6 | 0.7 | 0.1 | 0.02 | 4.0 | [34,134] |
| | Birmingham (USA) | 1.7 | 0.5 | 0.2 | 0.1 | 10.9 | [34,134] |
| | Atlanta (USA) | 0.8 | 0.3 | 0.2 | 0.01 | 6.2 | [34,134] |
| Europe | Cork Harbour (Ireland) | 0.02 | - | - | - | 1.1 | [135] |
| | Julich (Germany) | 0.1 | 0.1 | - | - | 4.4 | [136] |
| | Marseille (France) | 0.02 | 0.1 | - | 0.01 | 4.1 | [127] |
| | K-puszta (Hungary) | 0.3 | 0.2 | - | - | 3.5 | [137] |
| Asia | Mumbai (India) | 0.01 | 0.04 | 0.01 | 0.03 | 4.4 | [138] |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | Nam Co Lake (Tibet) | 0.4 | 0.04 | 0.1 | 0.04 | 1.2 | [133] |
| | Hong Kong (China) | 0.3 | 1.4 | 0.3 | 0.1 | 5.1 | [139] |
| | Hong Kong (China) | 0.3 | 1.2 | 0.2 | 0.7 | 2.4 | [139] |
| | Dinghu (China) | 0.2 | - | - | - | 4.9 | [140] |
| | PRD (China) | 0.6 | 0.1 | 2.3 | 0.1 | 4.7 | [63] |
| | Shanghai (China) | 0.3 | 0.03 | 0.2 | 0.03 | 5.7 | [59] |
| | Shanghai (China) | 0.4 | 0.04 | 0.3 | 0.1 | 5.7 | [59] |
| | Chongming (China) | 0.03 | - | - | - | 9.9 | [140] |
| | Mount Tai (China) | 1.0 | 0.2 | - | 0.5 | 15.9 | [141-143] |
| | Changbai (China) | 0.3 | - | - | - | 4.5 | [140] |
| | Beijing (China) | 0.9 | 0.5 | 1.7 | 0.2 | 7.1 | [144] |
| | Beijing (China) | 1.3 | 0.5 | 1.5 | 0.2 | 5.8 | [144] |
| | Hokkaido (Japan) | 0.5 | 0.1 | - | 0.03 | 3.6 | [145] |

Black: urban; Blue: suburban; Green: Remote; Red: rural.

**Annex D. Positive matrix factorization (PMF) approach.**

Table SD1. List of the studies considered for the annual SOC contribution to $PM_{2.5}$ OC estimated using the PMF approach (filer based) for all the monitored sites from 2006 to 2016. SOC and OC concentrations in $\mu gC\ m^{-3}$.

| **Locations** | | **SOC** | **OC** | **SOC contribution (%)** | **Molecular markers used for SOC estimation** | **References** |
|---|---|---|---|---|---|---|
| North America | Yorkville (USA)[1] | 1.0 | 2.9 | 34 | - | [146] |
| | Rochester (USA)[2] | 0.04 | 0.4 | 12 | √ | [147] |
| | Pittsburgh (USA)[3] | 1.0 | 2.9 | 34 | √ | [148] |
| | Cincinnati (USA)[4] | 1.2 | 2.8 | 43 | √ | [149] |
| | Detroit (USA)[4] | 1.6 | 3.2 | 51 | √ | [149] |
| | East St. Louis (USA)[4] | 1.01 | 3.5 | 30 | √ | [149] |
| | East St. Louis (USA)[5] | 0.8 | 3.8 | 21 | √ | [150] |
| | East St. Louis (USA)[5] | 0.8 | 4.0 | 20 | √ | [151] |
| | Bondville (USA)[4] | 0.9 | 1.6 | 57 | √ | [149] |
| | Northbrook (USA)[4] | 1.0 | 2.4 | 43 | √ | [149] |
| | Riverside (USA)[6] | 1.3 | 3.3 | 39 | √ | [101] |
| | Central LA (USA)[6] | 1.5 | 3.9 | 39 | √ | [101] |
| | Centreville (USA)[1] | 0.7 | 2.8 | 26 | - | [146] |
| | Birmingham (USA)[1] | 0.9 | 4.3 | 21 | - | [146] |
| | Atlanta (USA)[1] | 1.0 | 4.2 | 23 | - | [33,146] |
| Asia | Shanghai (China)[6,7] | 2.5 | 6.8[7] | 37 | √ | [59] |
| | Hong Kong (China)[6*] | 6.8 | 10.4 | 66 | √ | [152] |
| | Hong Kong (China)[6**] | 0.7 | 2.9 | 24 | √ | [152] |

Black: urban; Blue: suburban; Green: Remote; Red: rural;

[1]SOC based on sulfate and nitrate factors;

[2]SOC based on isoprene + other SOA factors;

[3]SOC based on biogenic SOA factor;

[4]SOC based on isoprene + pinene + caryophyllene SOA factors;

[5]SOC based on anthropogenic SOA factor;

[6]SOC based on biogenic + anthropogenic SOA + sulfate and nitrate factors;

[7] both urban and suburban sites considered;

[*] Regional days;

[**] Clean days.

Table SD2. List of the studies considered for the spring-summer SOC contribution estimated using the PMF approach (AMS/ACSM based). SOC and OC concentrations in μgC $m^{-3}$.

| Locations | | SOC | OC | SOC contribution (%) | References |
|---|---|---|---|---|---|
| North + South America | Vancouver (Canada) | 1.3 | 3.4 | 37 | [153] |
| | Chebogue Pt. (Canada)[1] | 0.8 | 0.9 | 81 | [153] |
| | Boulder (USA) | 1.28 | 1.7 | 74 | [153] |
| | New York City (USA) | 2.3 | 4.2 | 64 | [153-155] |
| | Riverside (USA) | 3.2 | 5.5 | 58 | [153,154] |
| | Houston (USA) | 1.4 | 4.2 | 33 | [153] |
| | Storm Peak (USA)[1] | 0.4 | 0.5 | 67 | [153] |
| | Thompson Farm (USA)[1] | 2.2 | 3.1 | 68 | [153] |
| | Pinnacle Park (USA)[1] | 2.8 | 3.0 | 91 | [153] |
| | Mexico City (Mexico) | 1.5 | 13.3 | 33 | [153,154] |
| Europe | Mace Head (Ireland) | 2.8 | 5.3 | 52 | [156] |
| | **Essex (UK)** | 1.4 | 2.2 | 64 | [154] |
| | **Essex (UK)** | 0.8 | 1.3 | 60 | [154] |
| | **Chelmsford (UK)** | 0.9 | 1.6 | 56 | [153] |
| | Manchester (UK) | 1.5 | 4.6 | 33 | [153,154] |
| | Edinburgh (UK) | 0.6 | 2.3 | 27 | [153] |
| | Hyytiälä (Finland) | 3.9 | 4.5 | 87 | [156] |
| | Helsinki (Finland) | 3.0 | 5.2 | 57 | [156] |
| | Chilbolton (UK) | 2.5 | 5.5 | 46 | [156] |
| | Vavihill (Sweden) | 2.9 | 5.2 | 56 | [156] |
| | Cabauw (Netherlands) | 3.1 | 5.0 | 61 | [156] |
| | Payerne (Switzerland) | 3.5 | 4.7 | 75 | [156] |
| | Zurich (Switzerland) | 2.2 | 2.7 | 83 | [153] |
| | *Jungfraujoch (Switzerland)* | 3.5 | 4.7 | 74 | [156] |
| | **Taunus (Germany)** | 4.1 | 5.5 | 74 | [153] |
| | Melpitz (Germany) | 3.7 | 4.6 | 80 | [156] |

| | | | | | |
|---|---|---|---|---|---|
| | Paris (France) | 0.6 | 0.9 | 61 | [157] |
| | *Puy de Dome (France)* | 3.2 | 5.0 | 64 | [156] |
| | Barcelona (Spain) | 2.1 | 4.4 | 47 | [156] |
| | Montseny (Spain) | 3.5 | 4.7 | 75 | [156] |
| | San Pietro Capofiume (Italy) | 3.3 | 4.9 | 67 | [156] |
| | Finokalia (Greece) | 4.0 | 4.3 | 93 | [156] |
| Asia | Beijing (China) | 7.7 | 15.2 | 51 | [85,153,154,158,159] |
| | Shanghai (China) | 5.1 | 7.3 | 71 | [85,158] |
| | Lanzhou (China) | 3.5 | 7.7 | 45 | [158] |
| | Shenzhen (China) | 8.6 | 15.9 | 54 | [85] |
| | Jiaxing (China) | 3.7 | 6.7 | 55 | [160] |
| | **Back Garden (China)** | 3.8 | 7.3 | 52 | [158] |
| | **Jiaxing (China)** | 5.3 | 8.4 | 62 | [85,158] |
| | **Changdao (China)** | 4.1 | 7.3 | 56 | [158] |
| | **Hong Kong (China)** | 3.7 | 5.3 | 73 | [85,158] |
| | Tokyo (Japan) | 2.21 | 4.2 | 52 | [153,154] |
| | Fukue (Japan)[1] | 2.4 | 2.7 | 88 | [153,154] |
| | Okinawa Island (Japan)[1] | 2.2 | 2.2 | 100 | [154] |
| | Cheju (Korea)[1] | 0.2 | 0.5 | 43 | [153] |

Black: urban; Blue: suburban; Green: Remote; Red: rural; Italic: High altitude; Bold: Urban Downwind

[1] mentioned as Remote/ Rural, not clearly specified.

**Annex E. Radiocarbon ($^{14}C$) measurements.**

Table SE1. List of the studies considered for the spring-summer SOC contribution to $PM_{2.5}$ OC estimated $^{14}C$ measurements in combination with another methodology. SOC and POC concentrations in μgC $m^{-3}$.

| Locations | | $SOC_{contemporary}$ | $SOC_{fossil}$ | POC | Methodology used to estimate SOC | References |
|---|---|---|---|---|---|---|
| North America | Great Smoky Mountain (USA) | 0.7 | 0.1 | 1.1 | EC tracer method | [161] |
| | Brigantine (USA) | 0.6 | 0.1 | 1.0 | EC tracer method | [161] |
| | Proctor Maple (USA) | 0.6 | 0.02 | 0.9 | EC tracer method | [161] |
| | Sula (USA) | 0.4 | 0.0 | 0.6 | EC tracer method | [161] |
| | Puget Sound (USA) | 0.4 | 0.3 | 1.0 | EC tracer method | [161] |
| | Mount Rainier (USA) | 0.6 | 0.1 | 1.0 | EC tracer method | [161] |
| | Yosemite (USA) | 1.3 | 0.04 | 2.1 | EC tracer method | [161] |
| | Grand Canyon (USA) | 0.3 | 0.01 | 0.4 | EC tracer method | [161] |
| | Tonto (USA) | 0.3 | 0.04 | 0.5 | EC tracer method | [161] |
| | Phoenix (USA) | 0.5 | 0.2 | 1.0 | EC tracer method | [161] |
| | Rocky Mountain (USA) | 0.4 | 0.04 | 0.7 | EC tracer method | [161] |
| | Pensacola (USA) | 1.9 | 1.0 | 1.4 | CMB approach | [162] |
| | Centreville (USA) | 3.0 | 0.7 | 0.9 | CMB approach | [162] |
| | Birmingham (USA) | 2.3 | 2.6 | 2.3 | CMB approach | [162] |
| | Atlanta (USA) | 3.7 | 1.6 | 1.6 | CMB approach | [162] |
| Europe | Aveiro (Portugal) | 1.3 | 0.2 | 2.0 | Using empirical equations | [163] |
| | Marseille (France) | 2.0 | 0.6 | 0.8 | PMF-AMS & CMB approaches | [127,164] |

| | | | | | | |
|---|---|---|---|---|---|---|
| | Puy de Dome (France) | 2.0 | 0.3 | 2.4 | Empirical equations | [163] |
| | Po Valley (Italy) | 3.4 | 1.3 | 1.2 | Empirical equations | [165] |
| | K-puszta (Hungary) | 1.8 | 0.1 | 2.6 | Empirical equations | [163] |
| | Sonnblick (Austrian Alps) | 0.6 | 0.1 | 0.8 | Empirical equations | [163] |
| | Schauinsland (Germany) | 1.5 | 0.3 | 2.0 | Empirical equations | [163] |
| | Goteborg (Sweden) | - | 0.5 | 1.8 | WSOC based method | [166] |
| Asia | Beijing (China) | 3.7 | 3.9 | 7.7 | Simulations and empirical equations | [167,168] |
| | Guangzhou (China) | 4.1 | 2.0 | 4.2 | Empirical equations | [167] |

Black: urban; Blue: suburban; Green: Remote; Red: rural;

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