

SUPPLEMENTARY MATERIAL

Fossil versus contemporary sources of fine elemental and organic carbonaceous particulate matter during the DAURE campaign in Northeast Spain

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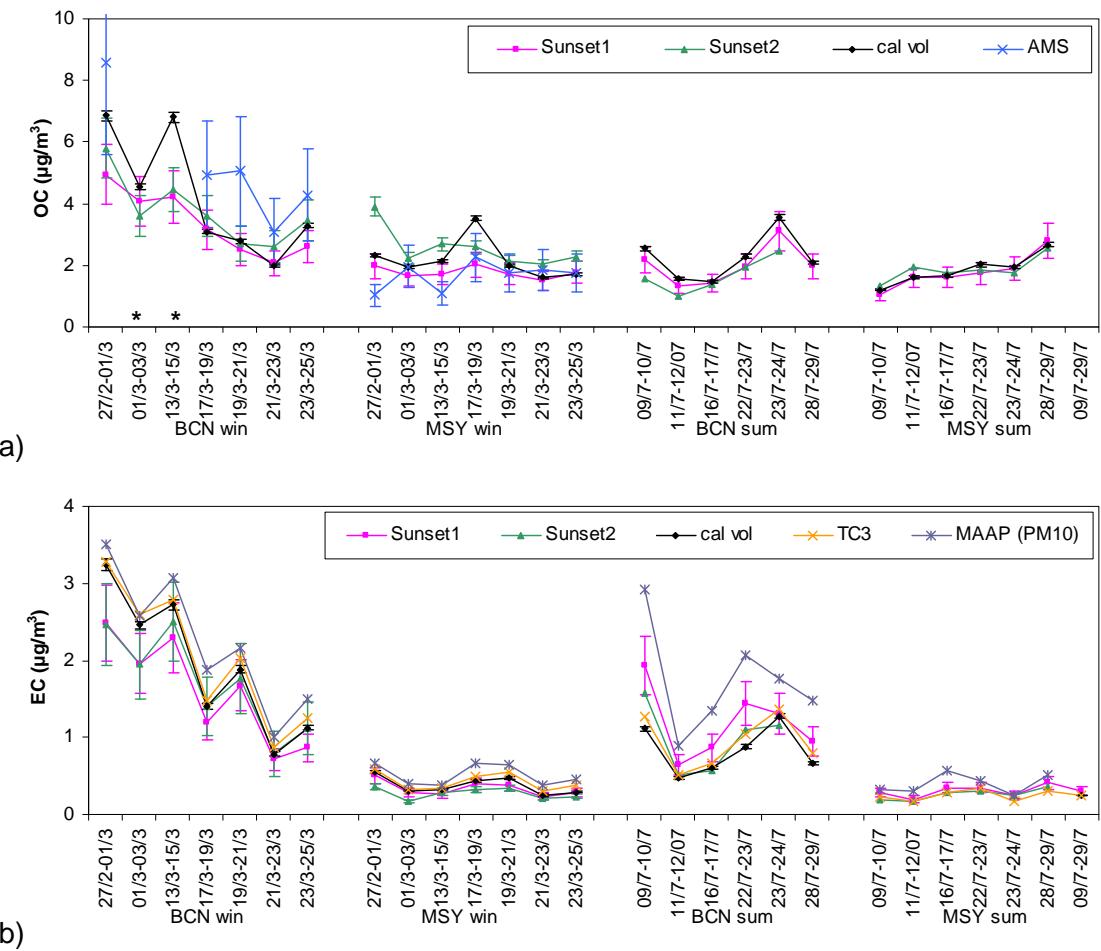


Figure S1. a) Organic carbon (OC) concentrations in PM₁ measured by Sunset1, Sunset2 and calibrated volume (cal vol); and calculated from organic matter measured by AMS using the AMS-determined OM/OC ratios of the different OA components for BCN and using the OM/OC ratios determined by AMS for MSY . *: Low availability of AMS data, so average not plotted.

b) Elemental carbon (EC) concentrations in PM₁ measured by Sunset1, Sunset2 and calibrated volume (cal vol); measured by the Sunset instrument during the third stage of thermal program for EC collection (TC3); and measured by MAAP with PM₁₀ inlet.

Table S1. Compilation of literature values of the EC/OC and levoglucosan/OC ratios for biomass burning emissions. SW: softwood; HW hardwood.

| Reference | Notes | (EC/OC) _{bb} | (EC/OC) _{bb} | (lev/OC) _{bb} | (lev/OC) _{bb} |
|--------------------------|-------------------------|-----------------------|-----------------------|------------------------|------------------------|
| Bond et al., 2004 | | 0.161 | | | |
| Chow et al., 2010 | residential wood comb | 0.188 | | | |
| Chow et al., 2010 | agricultural burning | 0.300 | | | |
| Chow et al., 2010 | open/prescribed burning | 0.180 | | | |
| Chow et al., 2010 | forest fire | 0.043 | | | |
| Chow et al., 2010 | wildfires | 0.513 | | | |
| Chow et al., 2010 | fires | 0.513 | | | |
| Fine et al. (2002, 2004) | Pine | | | 0.258 | 0.052 |
| Fine et al. (2002, 2004) | Region 5 profile | | | 0.146 | 0.029 |
| Fine et al. (2002, 2004) | Region 4 profile | | | 0.093 | 0.019 |
| Fine et al., 2004 | Quaking Aspen | 0.016 | 0.003 | 0.188 | |
| Fine et al., 2004 | Douglas Fir | 0.054 | 0.007 | 0.271 | |
| Fine et al., 2004 | Ponderosa Pine | 0.081 | 0.009 | 0.071 | |
| Fine et al., 2004 | Pinyon Pine | 0.408 | 0.032 | 0.01 | |
| Fine et al., 2004 | White Oak | 0.015 | 0.003 | 0.098 | |
| Fine et al., 2004 | Sugar Maple | 0.044 | 0.006 | 0.168 | |
| Fine et al., 2004 | Black Oak | 0.030 | 0.004 | 0.234 | |
| Fine et al., 2004 | American Beech | 0.015 | 0.003 | 0.076 | |
| Fine et al., 2004 | Black Cherry | 0.022 | 0.006 | 0.334 | |
| Fine et al., 2004 | White Spruce | 0.039 | 0.005 | 0.142 | |
| Gonçalves et al., 2010 | Eucalyptus globulus HW | 0.32 | | 0.462 | |
| Gonçalves et al., 2010 | Pinus pinaster SW | 1.11 | | 0.146 | |
| Gonçalves et al., 2010 | Quercus suber HW | 0.23 | | 0.159 | |
| Gonçalves et al., 2010 | Acacia longifolia HW | 1 | | 0.096 | |
| Liousse et al., 1996 | | 0.139 | | | |
| McMeeking et al., 2009 | Montane | 0.022 | | | |
| McMeeking et al., 2009 | Douglas fir | 0.014 | | | |
| McMeeking et al., 2009 | Lodgepole pine | 0.040 | | | |
| McMeeking et al., 2009 | Ponderosa pine | 0.027 | | | |
| McMeeking et al., 2009 | Rangeland | 0.128 | | | |
| McMeeking et al., 2009 | Juniper | 3.857 | | | |
| McMeeking et al., 2009 | Rabbitbrush | 2.800 | | | |
| McMeeking et al., 2009 | Sagebrush | 0.041 | | | |
| McMeeking et al., 2009 | Chaparral | 0.076 | | | |
| McMeeking et al., 2009 | Ceanothus | 0.092 | | | |
| McMeeking et al., 2009 | Chamise | 0.175 | | | |
| McMeeking et al., 2009 | Manzanita | 0.024 | | | |
| McMeeking et al., 2009 | Coastal plain | 0.073 | | | |
| McMeeking et al., 2009 | Black needlerush | 0.016 | | | |
| McMeeking et al., 2009 | Common reed | 0.020 | | | |
| McMeeking et al., 2009 | Gallberry | 1.141 | | | |
| McMeeking et al., 2009 | Hickory | 0.042 | | | |
| McMeeking et al., 2009 | Kudzu | 0.014 | | | |
| McMeeking et al., 2009 | Longleaf pine | 0.039 | | | |
| McMeeking et al., 2009 | Oak | 0.038 | | | |
| McMeeking et al., 2009 | Palmetto | 0.094 | | | |
| McMeeking et al., 2009 | Rhododendron | 0.095 | | | |
| McMeeking et al., 2009 | Sawgrass | 0.120 | | | |
| McMeeking et al., 2009 | Turkey oak | 0.043 | | | |
| McMeeking et al., 2009 | Wax myrtle | 0.056 | | | |
| McMeeking et al., 2009 | Wire grass | 0.086 | | | |

| Reference | Notes | EC/OC | unc EC/OC | lev/OC | unc lev/OC |
|-------------------------|----------------------------|--------------|----------------------|---------------|-----------------------|
| McMeeking et al., 2009 | Boreal forest | 0.026 | | | |
| McMeeking et al., 2009 | Alaskan duff | 0.000 | | | |
| McMeeking et al., 2009 | Black spruce | 0.097 | | | |
| McMeeking et al., 2009 | White spruce | 0.037 | | | |
| McMeeking et al., 2009 | Other | 160 | | | |
| McMeeking et al., 2009 | Fern | 0.045 | | | |
| McMeeking et al., 2009 | Rice straw | 194 | | | |
| Saarikoski et al., 2008 | | 0.152 | | | |
| Saarnio et al., 2010 | plume measurements | 0.100 | 0.068 | | |
| Saarnio et al., 2010 | plume measurements | 0.106 | 0.044 | | |
| Saarnio et al., 2010 | plume measurements | 0.149 | 0.068 | 0.028 | 0.014 |
| Sandradewi et al., 2008 | | 0.137 | | | |
| Schmidl et al., 2008 | Beech | 0.372 | | 0.080 | |
| Schmidl et al., 2008 | Oak | 0.308 | | 0.273 | |
| Schmidl et al., 2008 | Spruce | 0.384 | | 0.199 | |
| Schmidl et al., 2008 | Larch | 0.176 | | 0.272 | |
| Schmidl et al., 2008 | Briquettes | 0.760 | | 0.248 | |
| Sullivan et al., 2008 | Alaskan Duff | | | 0.117 | |
| Sullivan et al., 2008 | Black Needle Rush | 0.039 | | 0.080 | |
| Sullivan et al., 2008 | Black Spruce | 0.312 | | 0.073 | |
| Sullivan et al., 2008 | Black Spruce, dried | 0.087 | | 0.083 | |
| Sullivan et al., 2008 | Black Spruce, fresh | 0.010 | | 0.079 | |
| Sullivan et al., 2008 | Ceanothus | 0.098 | | 0.053 | |
| Sullivan et al., 2008 | Chamise | 0.668 | | 0.065 | |
| Sullivan et al., 2008 | Fir | | | 0.049 | |
| Sullivan et al., 2008 | Gallberry | 1.068 | | 0.033 | |
| Sullivan et al., 2008 | Grass | 0.050 | | 0.030 | |
| Sullivan et al., 2008 | Hickory | 0.043 | | 0.037 | |
| Sullivan et al., 2008 | Juniper | 3.455 | | 0.015 | |
| Sullivan et al., 2008 | Kudzo | | | 0.025 | |
| Sullivan et al., 2008 | Lodgepole Pine Needle Duff | | | 0.134 | |
| Sullivan et al., 2008 | Lodgepole Pine, dead/small | 1.042 | | 0.186 | |
| Sullivan et al., 2008 | Lodgepole Pine, fresh | 0.005 | | 0.053 | |
| Sullivan et al., 2008 | Longleaf Pine | 0.042 | | 0.072 | |
| Sullivan et al., 2008 | Manzanita | 0.293 | | 0.072 | |
| Sullivan et al., 2008 | Oak | 0.038 | | 0.062 | |
| Sullivan et al., 2008 | Palmetto | 0.364 | | 0.055 | |
| Sullivan et al., 2008 | Phragmites | 0.019 | | 0.075 | |
| Sullivan et al., 2008 | Ponderosa Pine Duff | 0.009 | | 0.069 | |
| Sullivan et al., 2008 | Ponderosa Pine, dead | 0.790 | | 0.102 | |
| Sullivan et al., 2008 | Ponderosa Pine, fresh | | | 0.070 | |
| Sullivan et al., 2008 | Puerto Rican Fern | 0.058 | | 0.070 | |
| Sullivan et al., 2008 | Puerto Rican Mixed Woods | 0.104 | | 0.129 | |
| Sullivan et al., 2008 | Rhododendron | 0.101 | | 0.101 | |
| Sullivan et al., 2008 | Rice Straw (Taiwan) | 0.039 | | 0.076 | |
| Sullivan et al., 2008 | Sage | 0.023 | | 0.028 | |
| Sullivan et al., 2008 | Saw Grass | 0.124 | | 0.041 | |
| Sullivan et al., 2008 | Southern Pine, dried | 0.068 | | 0.098 | |
| Sullivan et al., 2008 | Titi | 0.343 | | 0.051 | |
| Sullivan et al., 2008 | Turkey Oak | 0.045 | | 0.047 | |
| Sullivan et al., 2008 | Wax Myrtle | 0.131 | | 0.056 | |
| Sullivan et al., 2008 | Wax Myrtle | 0.038 | | 0.059 | |

| Reference | Notes | EC/OC | unc EC/OC | lev/OC | unc lev/OC |
|-----------------------|--------------|-------|--------------|--------|---------------|
| Sullivan et al., 2008 | White Spruce | 0.000 | | 0.133 | |
| Sullivan et al., 2008 | Wiregrass | 0.066 | | 0.201 | |
| Sullivan et al., 2008 | Wiregrass | 0.098 | | 0.172 | |
| Szidat et al., 2009 | | 0.157 | 0.05 | 0.1 | 0.08 |

Table 3 Szidat et al., 2006, JGR

| | | | | |
|-------------------------|-------|------|-------|------|
| Edgerton et al.1986 | 0.175 | | | |
| Rau, 1989 | 0.163 | | | |
| Hildemann et al.1991 | 0.084 | | | |
| Cachier et al.1996 | 0.116 | | | |
| Liousse et al.1996 | 0.234 | | | |
| McDonald et al.2000 | 0.182 | | | |
| Fine et al.2001 | 0.153 | | 0.103 | |
| Schauer et al.2001 | 0.04 | | 0.246 | |
| Fine et al.2002 | 0.159 | | 0.042 | |
| Fine et al. 2004a | 0.086 | | 0.134 | |
| Fine et al. 2004b | 0.213 | | 0.245 | |
| AVE Szidat et al., 2006 | 0.16 | 0.05 | 0.15 | 0.09 |

Table 2 Reid et al., 2005, ACP

| | | | | |
|----------------------|------------------|------|------|--|
| Andreae et al. 1998 | Savanna | 0.23 | | |
| Cachier et al. 1995 | Savanna | 0.12 | | |
| Cachier et al. 1996 | Savanna | 0.12 | | |
| Ferek et al. 1998 | Cerrado | 0.12 | | |
| Ferek et al. 1998 | Grass/Pasture | 0.11 | | |
| Formenti et al. 2003 | Savanna | 0.06 | 0.01 | |
| Liousse et al. 1995 | Savanna | 0.13 | 0.01 | |
| Ward et al. 1992 | Cerrado | 0.06 | | |
| Hobbs et al. 1996 | Presc. Temperate | 0.08 | 0.02 | |
| Mazurek et al. 1991 | Boreal | 0.07 | 0.02 | |
| Susott et al. 1991 | Temperate | 0.15 | | |
| Ward et al. 1992 | Temperate | 0.1 | | |
| Mazurek et al. 1991 | Boreal Forest | 0.08 | 0.03 | |
| Mazurek et al. 1991 | Boreal Forest | 0.03 | 0.03 | |
| Susott et al. 1991 | Temperate | 0.04 | | |
| Ferek et al. 1998 | S. Amer | 0.15 | | |
| Ferek et al. 1998 | S. Amer | 0.08 | | |
| Ward et al. 1992 | S. Amer | 0.2 | | |
| Patterson, 1984 | Forest Debris | 0.6 | | |
| Turn et al. 1997 | Herbaceous Fuel | 0.52 | | |
| Turn et al. 1997 | Woody Fuel | 0.48 | | |

References from Table S1:

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Sullivan, A.P., Holden, A.S., Patterson, L.A., McMeeking, G.R., Kreidenweis, S.M., Malm, W.C., Hao, W.M., Wold, C.E., and Collett, J.L.: A method for smoke marker measurements and its potential application for determining the contribution of biomass burning from wildfires and prescribed fires to ambient PM2.5 organic carbon, *J. Geophys. Res.-Atmos.*, 113, D22302, doi: 10.1029/2008JD010216, 2008.

Szidat, S., Jenk, T. M., Synal, H.-A., Kalberer, M., Wacker, L., Hajdas, I., Kasper-Giebl, A., and Baltensperger, U.: Contributions of fossil fuel, biomass-burning, and biogenic emissions to carbonaceous aerosols in Zurich as traced by ^{14}C . *J. Geophys. Res.-Atmos.*, 111, D07206, doi:10.1029/2005JD006590, 2006.

Szidat, S., Ruff, M., Perron, N., Wacker, L., Synal, H. A., Hallquist, M., Shannigrahi, A. S., Yttri, K. E., Dye, C., and Simpson, D.: Fossil and non-fossil sources of organic carbon (OC) and elemental carbon (EC) in Göteborg, Sweden, *Atmos. Chem. Phys.*, 9, 1521-1535, 2009.

Section S1:

New method for EC collection

The new method for EC collection for correct ^{14}C determination (still under development, Zhang et al., 2011) consists of coupling a Sunset Instrument to the cryo-trap system (as opposed to an oven with a fixed temperature with the old method), so that the thermal cycles can be defined accurately. The whole thermal program is carried out under pure oxygen. The CO_2 evolving from the ‘EC step’ is trapped for subsequent ^{14}C analysis. The filter transmittance is monitored continuously by the attenuation of a laser, which is used to calculate the EC yields in the CO_2 collection stage. The thermal program used consists on a first step at 400°C (T1) during 150 seconds (including pre-step 1), a cooling time of 18 seconds, a second step at 500°C (T2) during 75 seconds, another cooling during 40 seconds, and a third step at 760°C (T3) during 150 seconds, named ‘EC step’, during which the CO_2 evolved is collected. All these temperatures are the set theoretical temperatures with the real thermal evolution being different (Figure S2). The second step in this method should be optimized with the goal of a complete removal of the carbonaceous interfering fraction with the best possible recovery of the elemental carbon. To this end, different temperatures for this second step (T2) need to be tested and results evaluated (Figure S2).

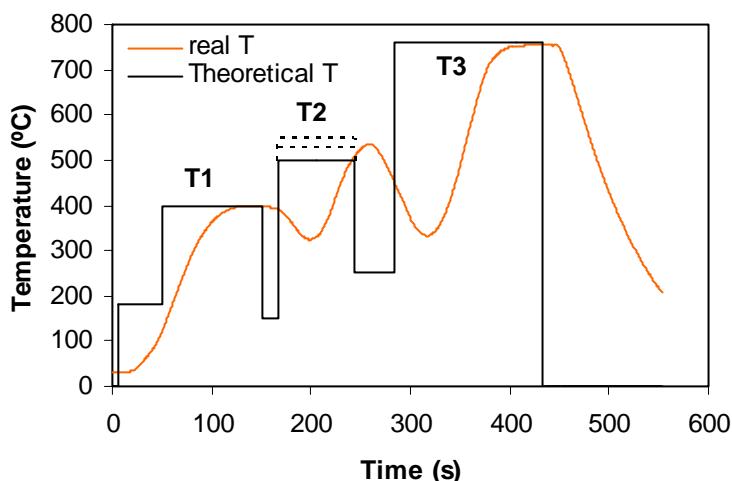


Figure S2. Thermal program on the Sunset instrument for the EC collection for subsequent ^{14}C analysis during the third step (at T3), showing the variation of T2 for optimizing tests.

References from Section S1:

- Zhang, Y.L., Perron, N., Zotter, P., Minguillón, M.C., Prévôt, A.S.H., Wacker, L., and Szidat, S.: On the isolation of organic carbon and elemental carbon of carbonaceous aerosols for ^{14}C measurement: A modified thermal/optical method, in preparation, 2011.

Section S2:

Selection of T2 for the EC collection thermal program

Different temperatures ranging from 485 to 600°C (as theoretical T2 temperatures) were tested using two filters from the study campaign, one from Barcelona (S5452) and one from Montseny (S5943). The yields of EC in the third step are defined by the attenuation (ATN) of the laser signal, hence the ratio ATN/ATNi (i standing for initial) indicates the amount of EC with respect to the initial (total) amount of EC in the filter. Figure S3 shows the ATN/ATNi right before step 3 for different T2. The EC yield does not change significantly when increasing the T2 up to around 560°C (real temperature). That indicates that the amount of carbon removed at the second step does not change, hence, any temperature chosen for T2 in this range would be valid, without any further information.

Additionally, analyses of fM of the collected CO₂ during the third step were carried out for different tests using different T2 (Figure S4).

The filter from Barcelona shows a slight decrease in the fM when increasing the T2, nevertheless the EC yield does not change, as explained above, this leading to the conclusion that the variation in the fM is not significant.

The filter from Montseny does not show any trend in the fM when increasing the T2 from 530 to 560 °C, and the EC yield didn't change significantly, as explained above.

Hence, for the type of samples of this study, the thermal program used allows us to get EC yields over 80% and the fM obtained from these yields is consistent and therefore it is considered representative of the EC collected and one can assume that it is representative of the whole EC fraction. The selection of the T2 could then be any T within the tested range, and the T2 was then set at 500°C (theoretical temperature, corresponding to 530°C as real temperature).

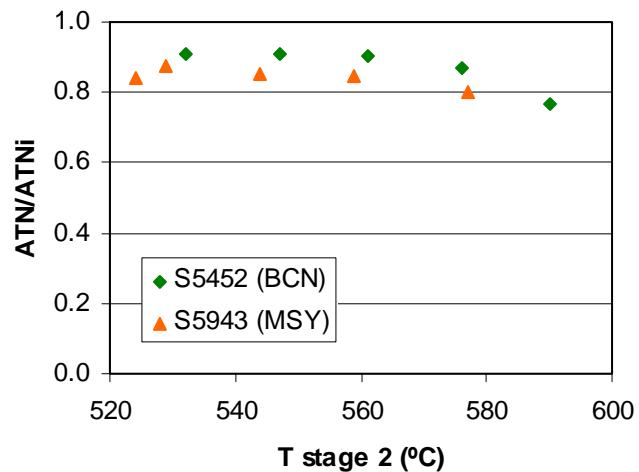


Figure S3. EC yields (ATN/ATNi) in the third step, defined as the attenuation of the laser signal after stage 2 and right before stage 3 (ATN) with respect to the initial attenuation of the laser signal (ATNi) for two filters depending on the temperature during stage 2.

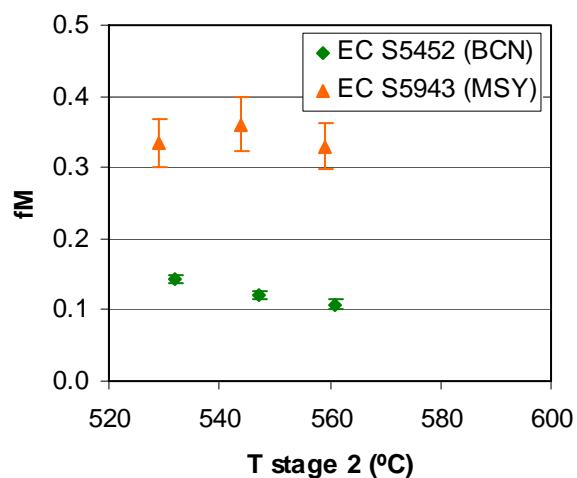


Figure S4. fM of collected CO₂ in the third step for two filters depending on the temperature during stage 2.

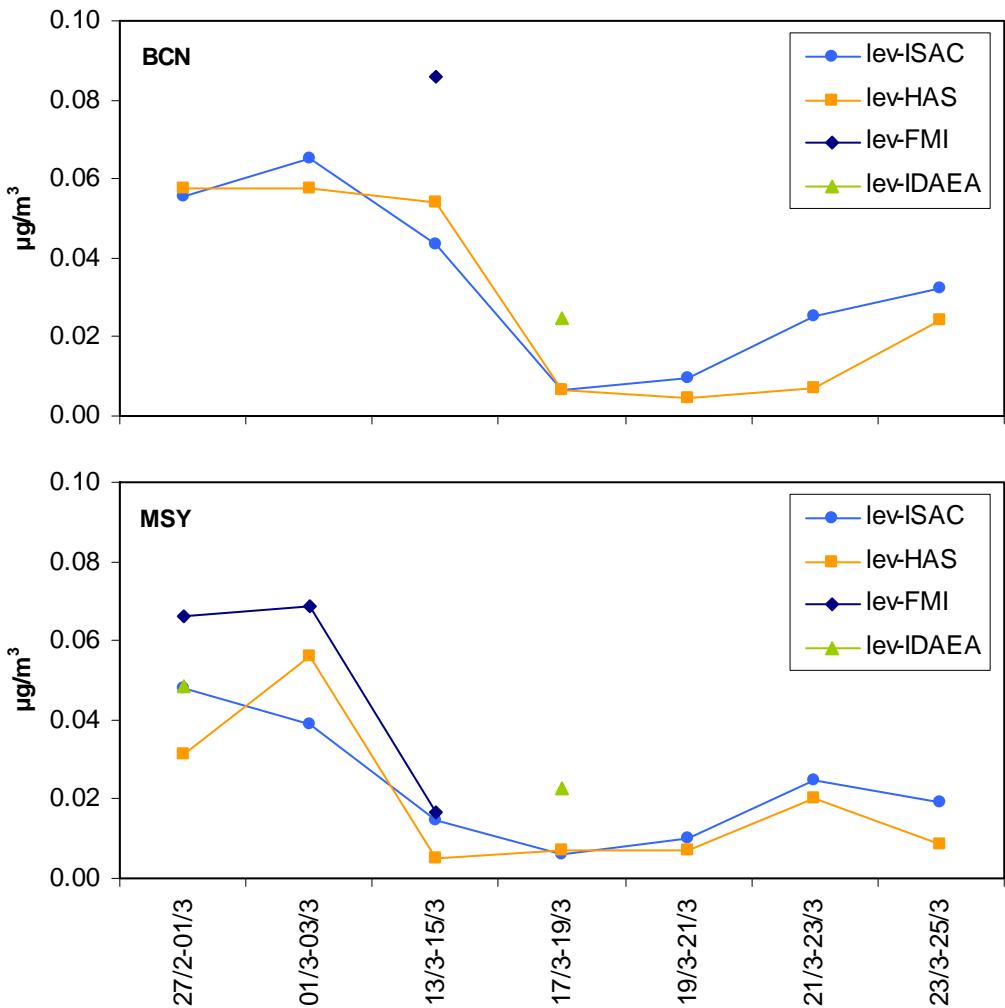
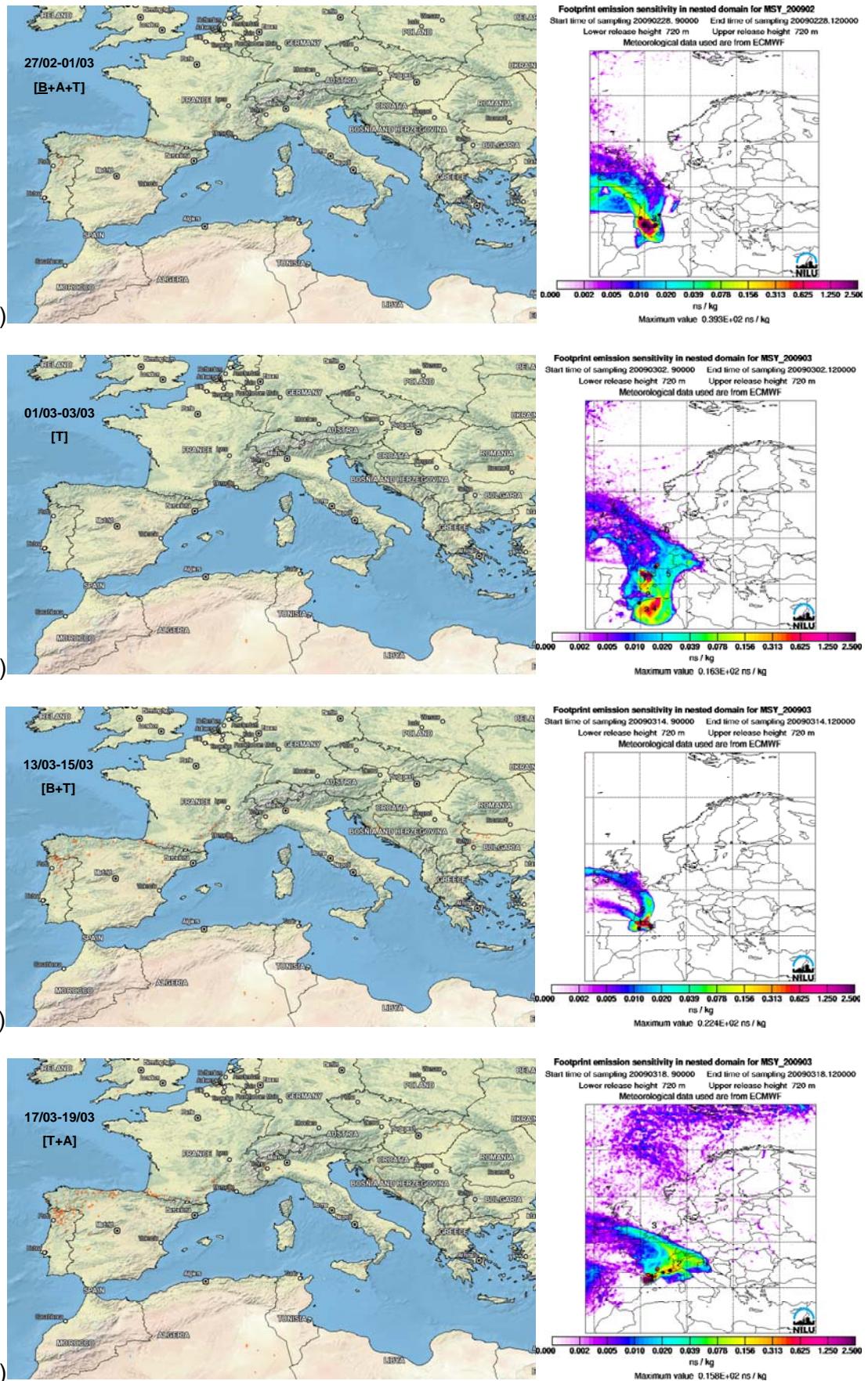
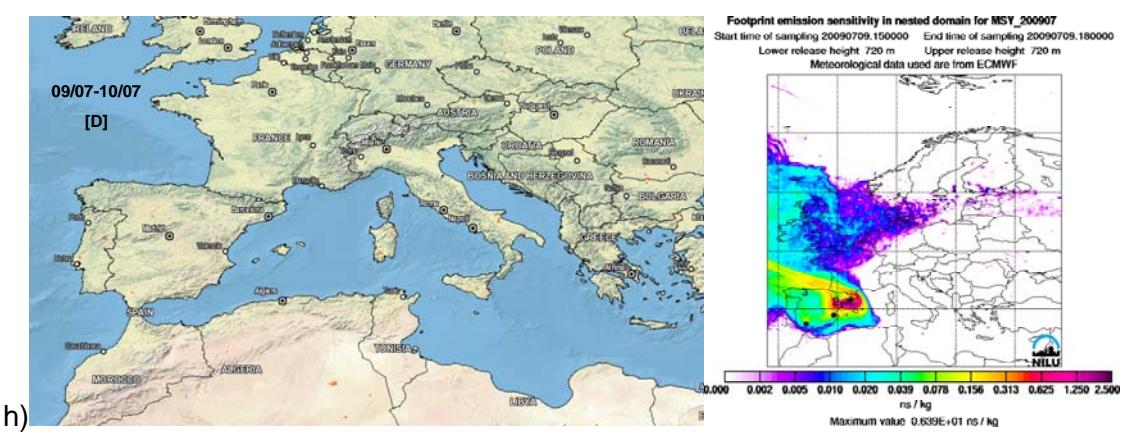
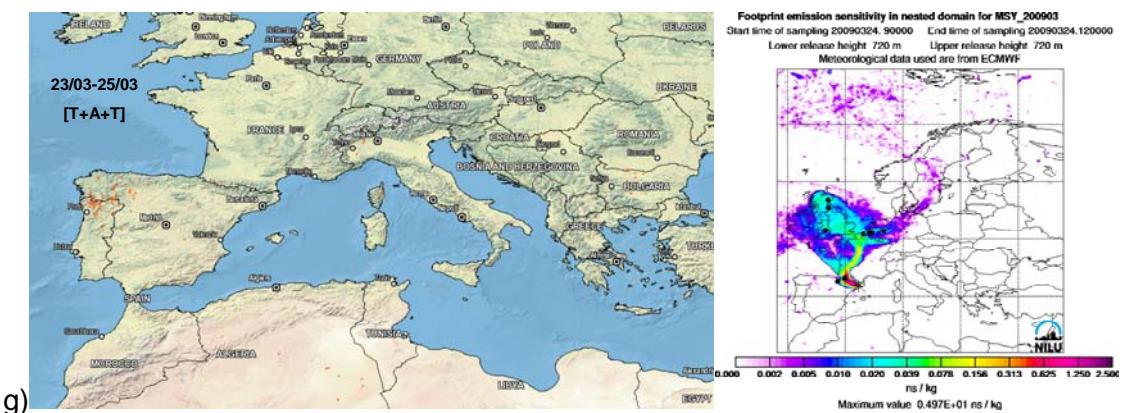
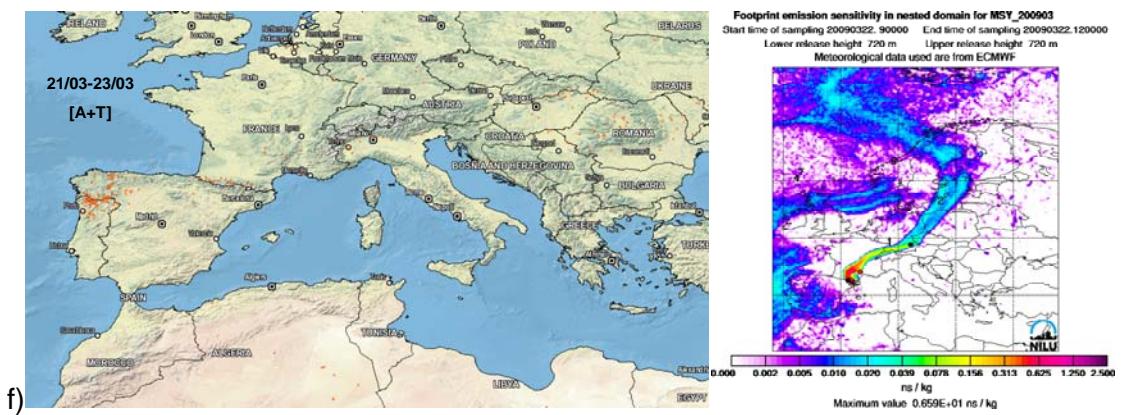
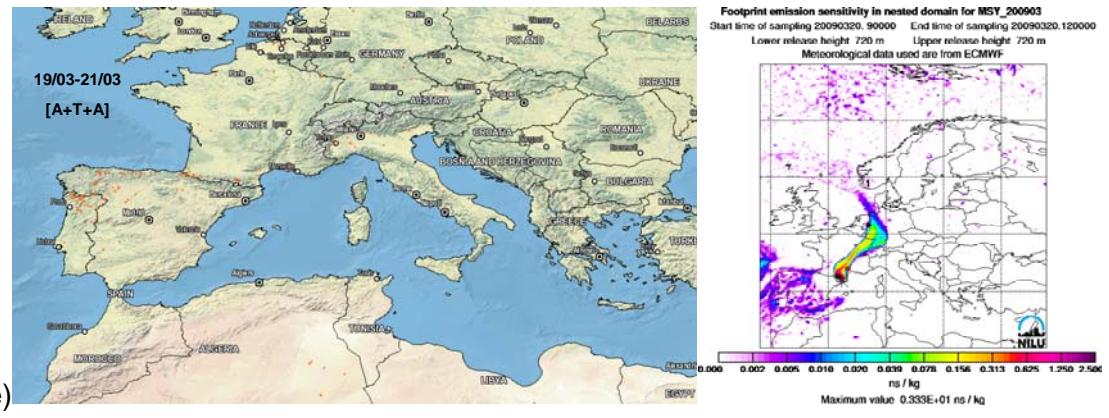
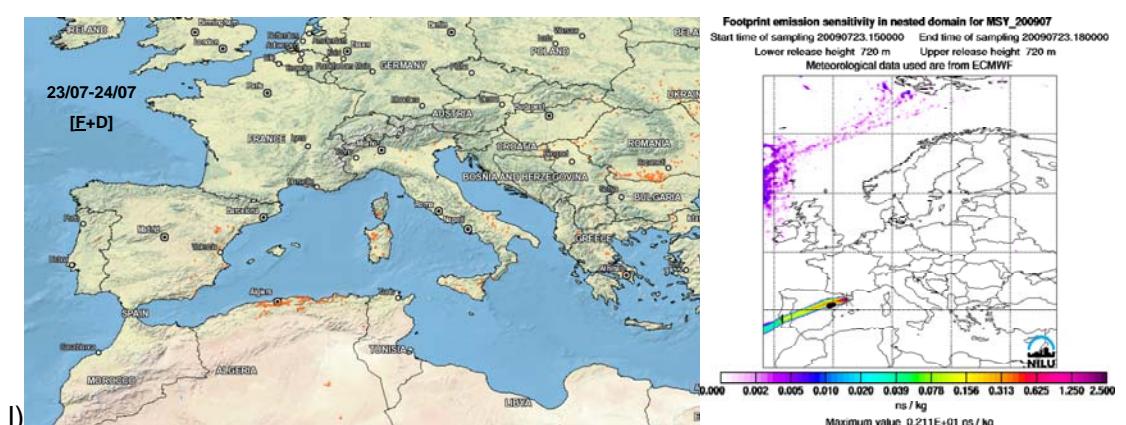
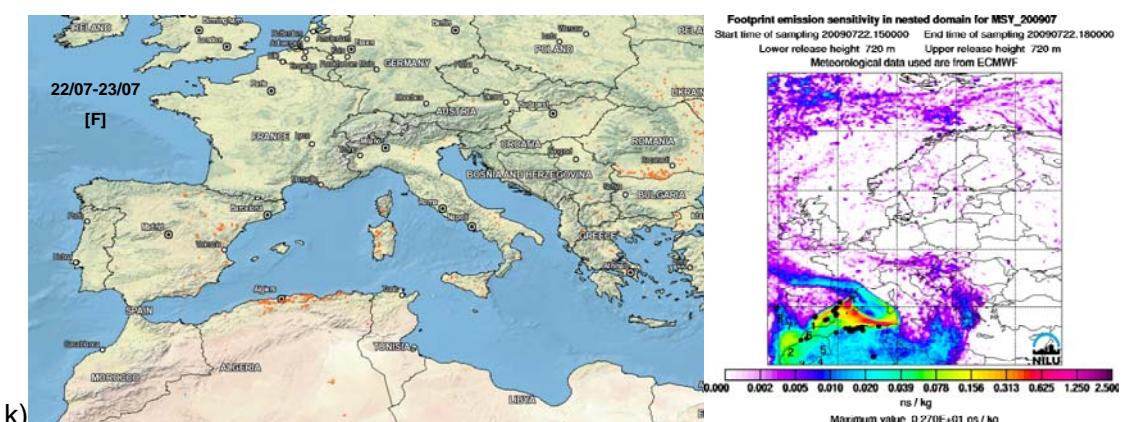
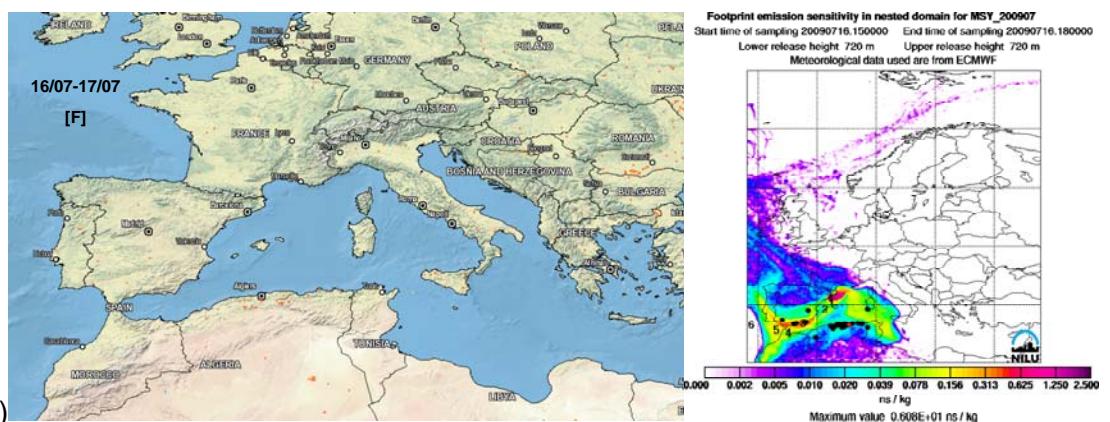
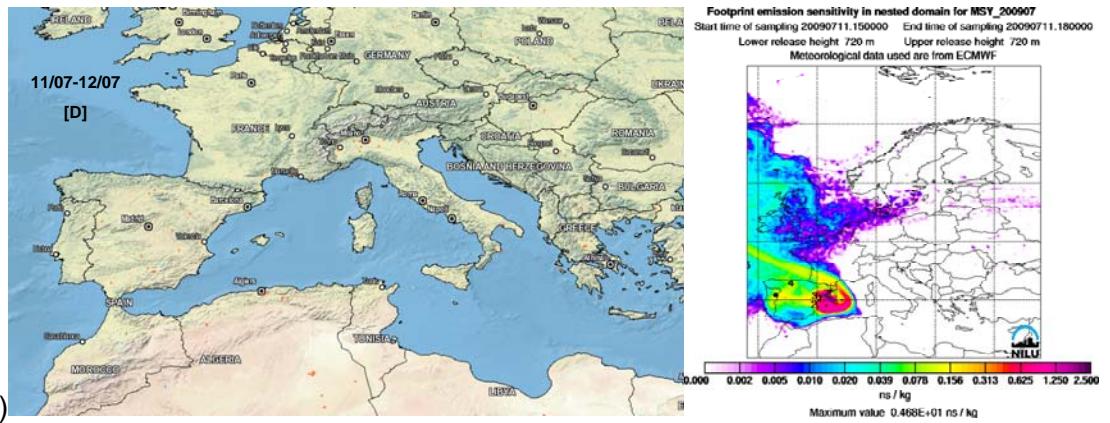
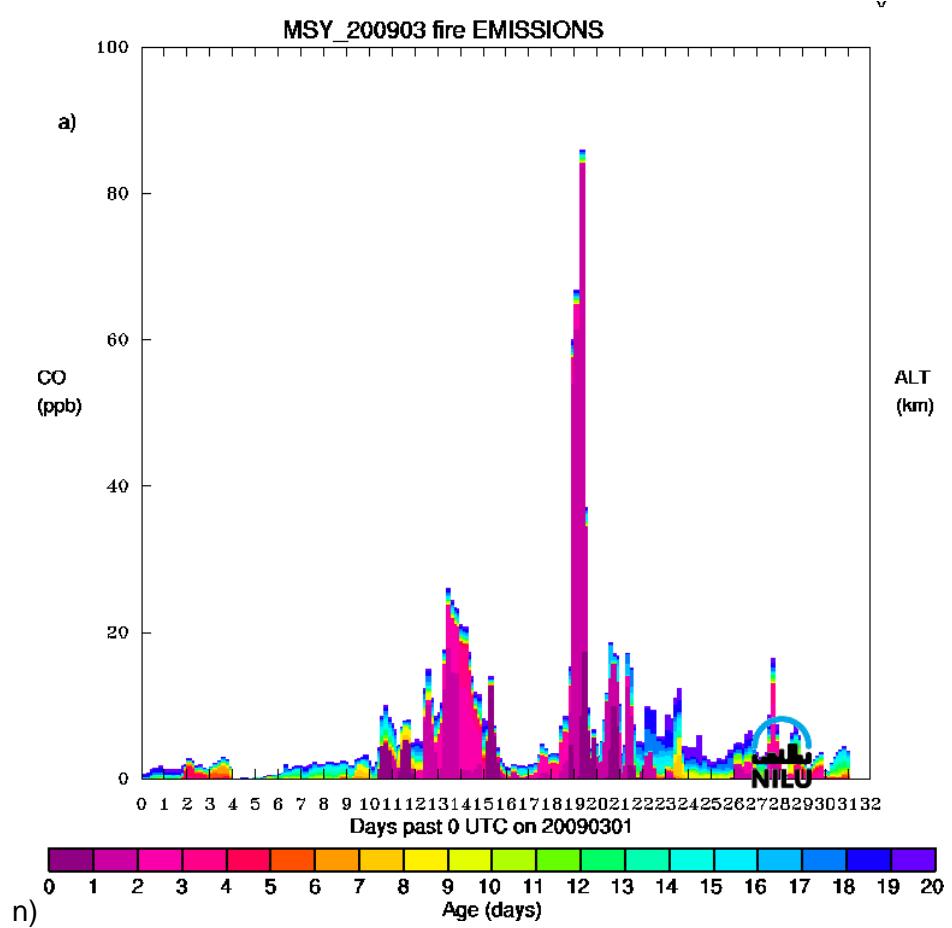
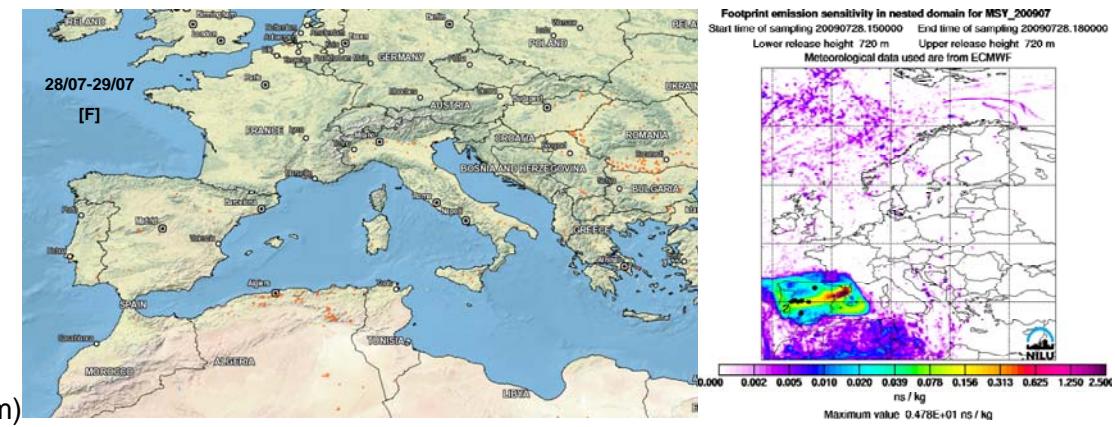


Figure S5. Levoglucosan concentrations determined by four different laboratories (see text for details). When the samples analyzed had a different time-resolution than the ^{14}C sampling periods (48h), averages of the corresponding samples are shown to match the 48h sampling periods. Only data matching the ^{14}C dataset period are shown, although more data from lev-FMI and lev-IDAEA are available.









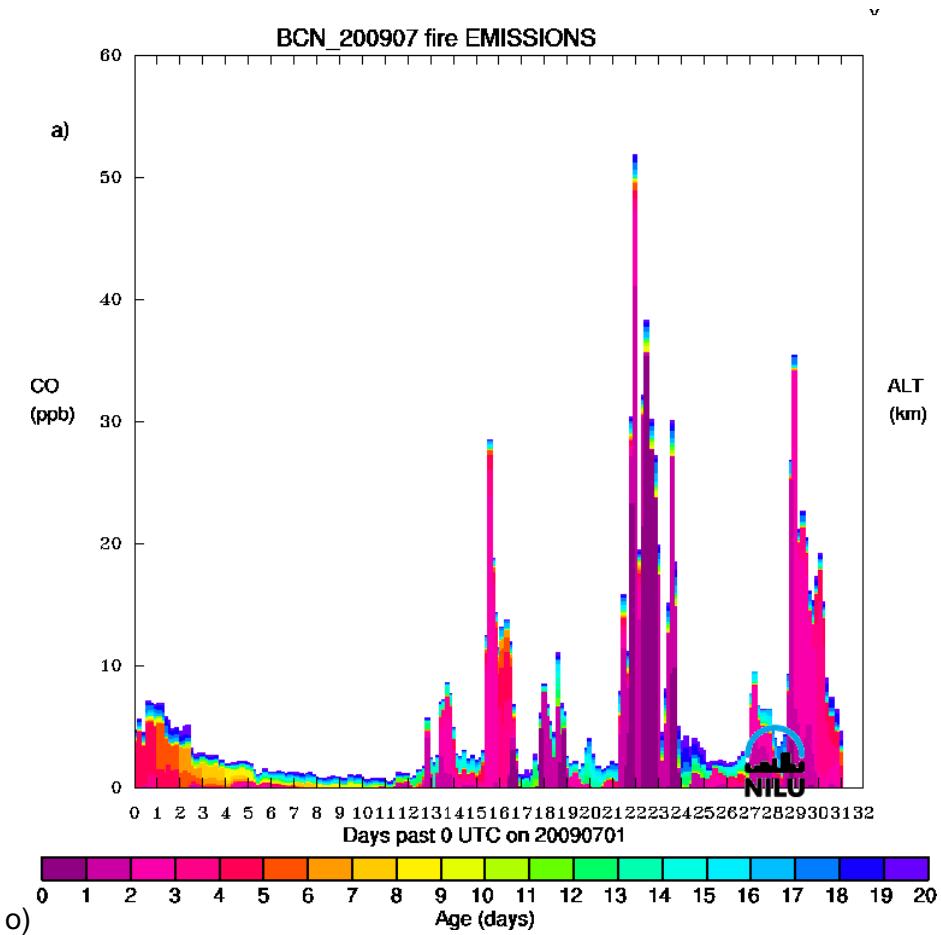


Figure S6. Hotspot/fire locations detected by the MODIS Rapid Response System provided by Web Fire Mapper, Fire Information for Resource Management System (FIRMS, Justice et al., 2002). Each map corresponds to a ^{14}C sample period (DAURE-W, a-g); DAURE-S, h-m). The atmospheric scenarios are listed below the dates; the prevailing scenario is underlined when more than one occurred during the same sampling period. FLEXPART air-mass sensitivities (Stohl et al., 2005) for MSY calculated for the middle of the sampling period. Wild fires impact estimated with FLEXPART during DAURE-W (n) and DAURE-S (o).

References from Figure S6:

Justice, C.O., Giglio, L., Korontzi, S., Owens, J., Morisette, J.T., Roy, D., Descloitres, J., Alleaume, S., Petitcolin, F., and Kaufman, Y.: The MODIS fire products, *Remote Sensing of Environment* 83, 244-262, 2002.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, *Atmos. Chem. Phys.*, 5, 2461-2474, 2005

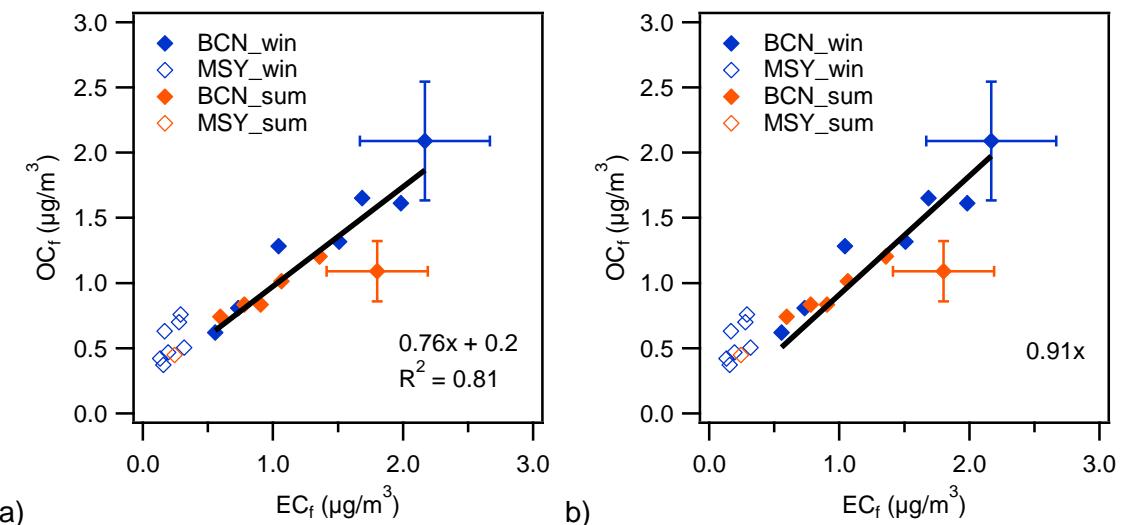


Figure S7. OC_f vs EC_f concentrations at BCN and MSY during DAURE-W and DAURE-S. Error bars indicate measurement uncertainty, only shown for two data points for clarity. Line and equation correspond to orthogonal distance regression for BCN. a) Regression line allowing intercept. b) Regression line with intercept forced to zero.

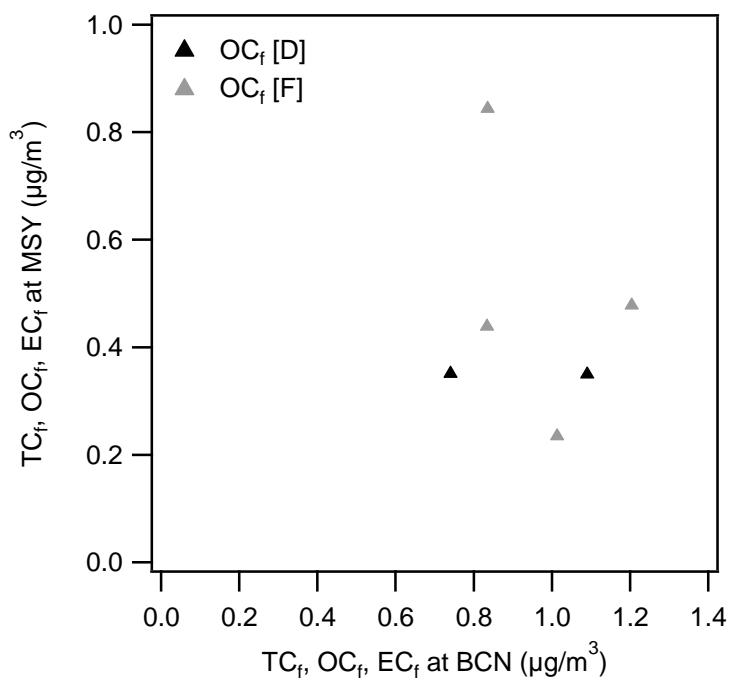


Figure S8. OC_f contributions at MSY vs OC_f contributions at BCN during scenario [D] and during scenario [F].