



Supplement of

Aerosol source apportionment modelling using a coupled regional–urban scale system

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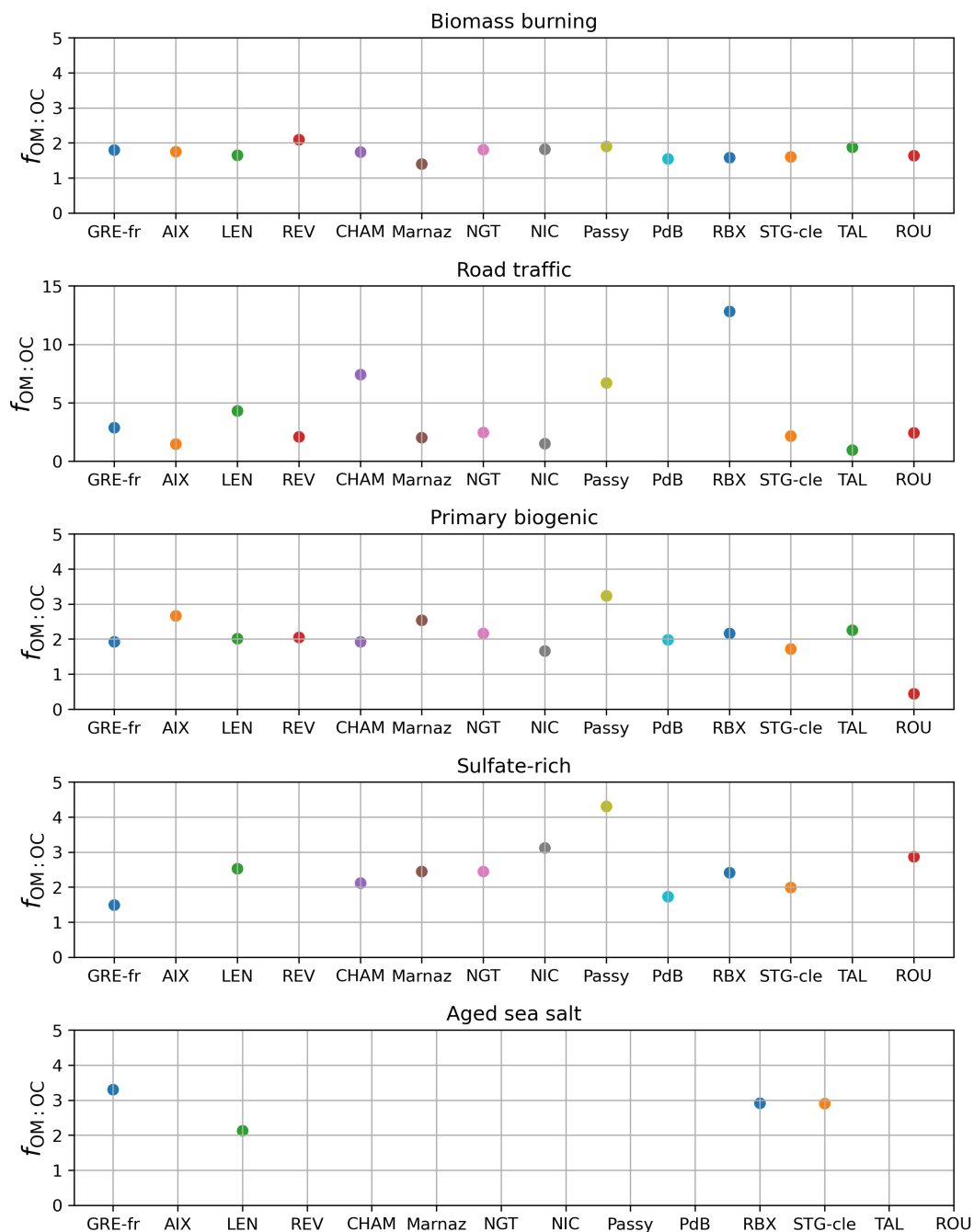


Figure S1. Derived OM to OC ratios ($f_{OM:OC}$) at each of the 13 Weber *et al.* (2019) sites for the biomass burning, road traffic, primary biogenic, sulfate-rich and aged sea salt PMF factors. Note the difference in y-axis scaling for the second panel from the top. Also note that not all PMF factors are included in the analysis at each of the stations.

Table S1. Long names, abbreviations, and latitude longitude (degrees) coordinates for all stations discussed in the main text.

| Station name | Abbreviation | Typology | Latitude | Longitude |
|----------------------|--------------|------------------|-------------|-------------|
| Barcelonne PR | BCN | Urban background | 41.38736 | 2.11544 |
| Basel, St. Johann | Basel | Urban background | 47.56591 | 7.58207 |
| Bern, Bollwerk | Bern | Traffic | 46.95099 | 7.44087 |
| Magadino, Cadenazzo | Mag | Rural | 46.16038 | 8.93394 |
| Payerne, MétéoSuisse | Paye | Rural | 46.81306 | 6.94447 |
| Zurich, Kaserne | Zurich | Urban background | 47.37759 | 8.53041 |
| Aix-en-provence | AIX | Urban background | 43.5302 | 5.4413 |
| Grenoble | GRE-fr | Urban background | 45.1618 | 5.7356 |
| Chamonix | CHAM | Urban valley | 45.9225 | 6.8699 |
| Lens | LEN | Urban background | 50.4368 | 2.8267 |
| Revin | REV | Rural | 49.91667 | 4.64139 |
| Marnaz | Marnaz | Urban valley | 46.0577 | 6.5334 |
| Nogent | NGT | Urban background | 49.2763 | 2.4821 |
| Nice | NIC | Urban traffic | 43.702 | 7.2862 |
| Passy | Passy | Urban valley | 45.9235 | 6.7136 |
| Port-de-Bouc | PdB | Industrial | 43.4019 | 4.9819 |
| Roubaix | RBX | Traffic | 50.7065 | 3.1806 |
| Rouen | ROU | Urban background | 49.42817 | 1.05808 |
| Strasbourg | STG-cle | Traffic | 48.5903 | 7.7450 |
| Talence | TAL | Urban background | 44.80056 | -0.58806 |
| Frauenfeld | fra | Urban background | 47.55608 | 8.89426 |
| Gallen | gal | Traffic | 47.42894 | 9.38653 |
| Vaduz | vad | Traffic | 47.12373 | 9.52361 |
| Vittore | vi | Rural | 46.23859 | 9.10505 |
| Lille | Lille | Urban | 50.611048 | 3.140368 |
| Paris | Paris | Urban | 48.708888 | 2.1426666 |
| London | London | Urban | 51.521046 | -0.2134921 |
| Marseille | Mar | Urban | 43.30530556 | 5.394694444 |
| Hohenpeissenberg | HPB | Rural | 47.80138889 | 11.00972222 |
| Puy de Dôme | Puy | Rural | 45.77222222 | 2.965833333 |

Table S2. Chemical composition of the lumped species discussed in the context of the Weber *et al.* (2019) PMF analysis. OC* is defined as the total OC content minus OC contributions from the individually measured organic molecules MSA, polyols, levoglucosan, and manosan, to avoid double-counting (Weber *et al.*, 2019).

| Lumped species | Included measured species |
|-------------------------------|--|
| OC | OC*, 0.12×MSA, 0.40×Polyols, 0.44×Levoglucosan, 0.44×Mannosan |
| EC | EC |
| Rest | Sb, Mn, Al, Co, Li, Ce, Tl, Cs, Fe, Cu, Na ⁺ , K ⁺ , Mg ²⁺ , Cl ⁻ , Cr, Sn, V, La, Rb, Ba, Mo, Ti, Se, As, Ni, Ca ²⁺ , Cd, Mn, Pb, Sr, Zn |
| NO ₃ ⁻ | NO ₃ ⁻ |
| SO ₄ ²⁻ | SO ₄ ²⁻ |
| NH ₄ ⁺ | NH ₄ ⁺ |

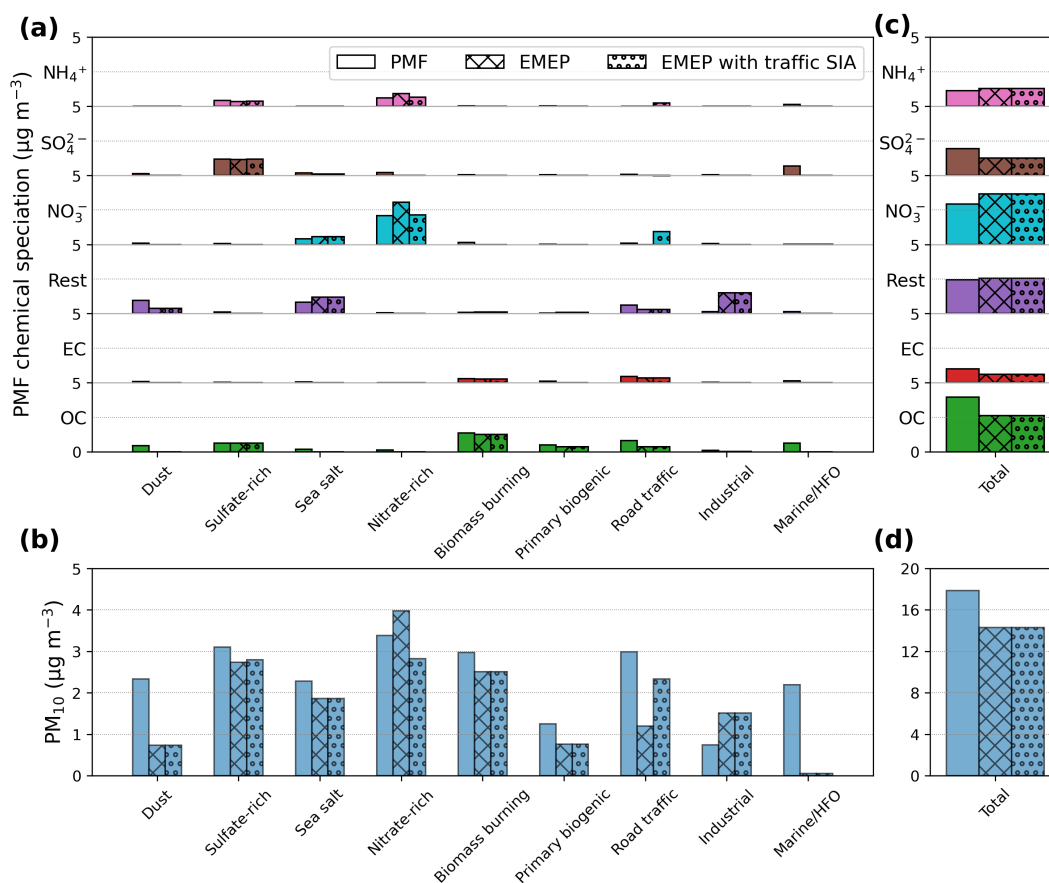


Figure S2. As Fig. 3 of the main text, but now comparing EMEP results to diagnostic EMEP calculations where SIA from on-road and off-road traffic is added to the ‘road traffic’ PMF factor calculations.

Table S3. Model matching for the Weber et al. (2021) PMF factors. ‘All modelled PPM’ from any given source refers to its EC, OA, and Rest-PPM components. A discussion of the PMF factors as derived from measurement data is given in the main text.

| Modelled PMF factor | Assigned model outputs |
|--------------------------|---|
| Biomass burning | All modelled PPM from GNFR C and forest fires |
| Road traffic | All modelled vehicular exhaust, tyre wear and brake wear PPM. All modelled anthropogenic SOA. All modelled GNFR I (off-road) PPM. |
| Primary biogenic aerosol | All modelled PBAPs. All modelled GNFR L (non-livestock agricultural activity). |
| Dust | All modelled natural (wind blown) dust. All modelled road traffic road abrasion. |
| Sea salt | All modelled sea salt. All modelled coarse-mode NO_3^- . |
| Nitrate-rich | All fine-mode NO_3^- minus the contribution from the modelled Industrial PMF factor. All modelled NH_4^+ as split between this and the sulfate-rich factor. |
| Sulfate-rich | All SO_4^{2-} minus the contribution from the modelled Industrial PMF factor. All modelled NH_4^+ as split between this and the nitrate-rich factor. Background OA. All biogenic SOA. |
| Marine/HFO | All modelled PPM and SIA (NH_4^+ , SO_4^{2-} , NO_3^-) from GNFR G (shipping), where the SIA contributions are tracked using the LFs. |
| Industrial | All modelled PPM from GNFR B (industry). |

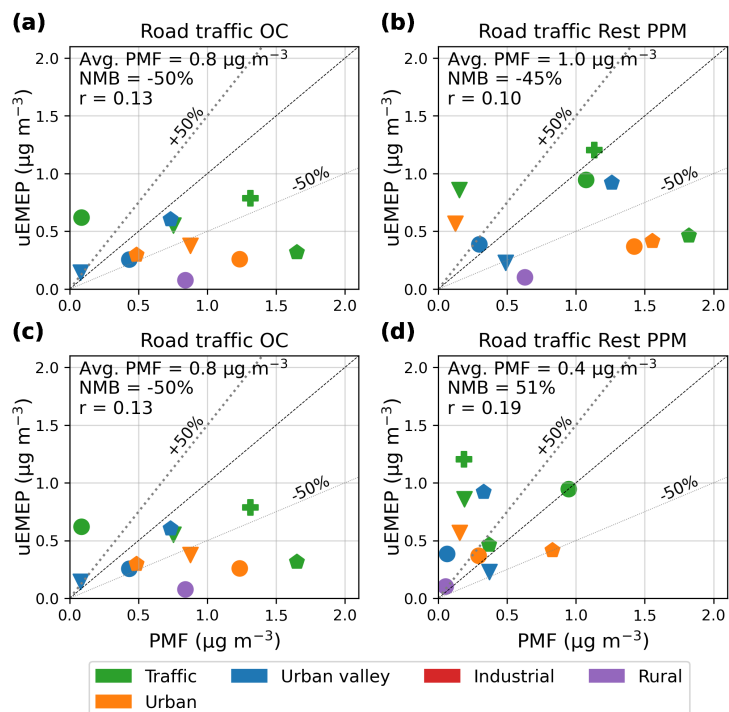


Figure S3. Comparison between PMF derived and modelled road traffic OC and Rest-PPM concentrations at the Weber et al. (2019) sites. Compared to Fig. 4 of the main text, panels (a) and (b) assume an alternative $f_{\text{OA:OC}}$ limit ratio of 1.4 while panels (c) and (d) assume an alternative ratio of 2.9.

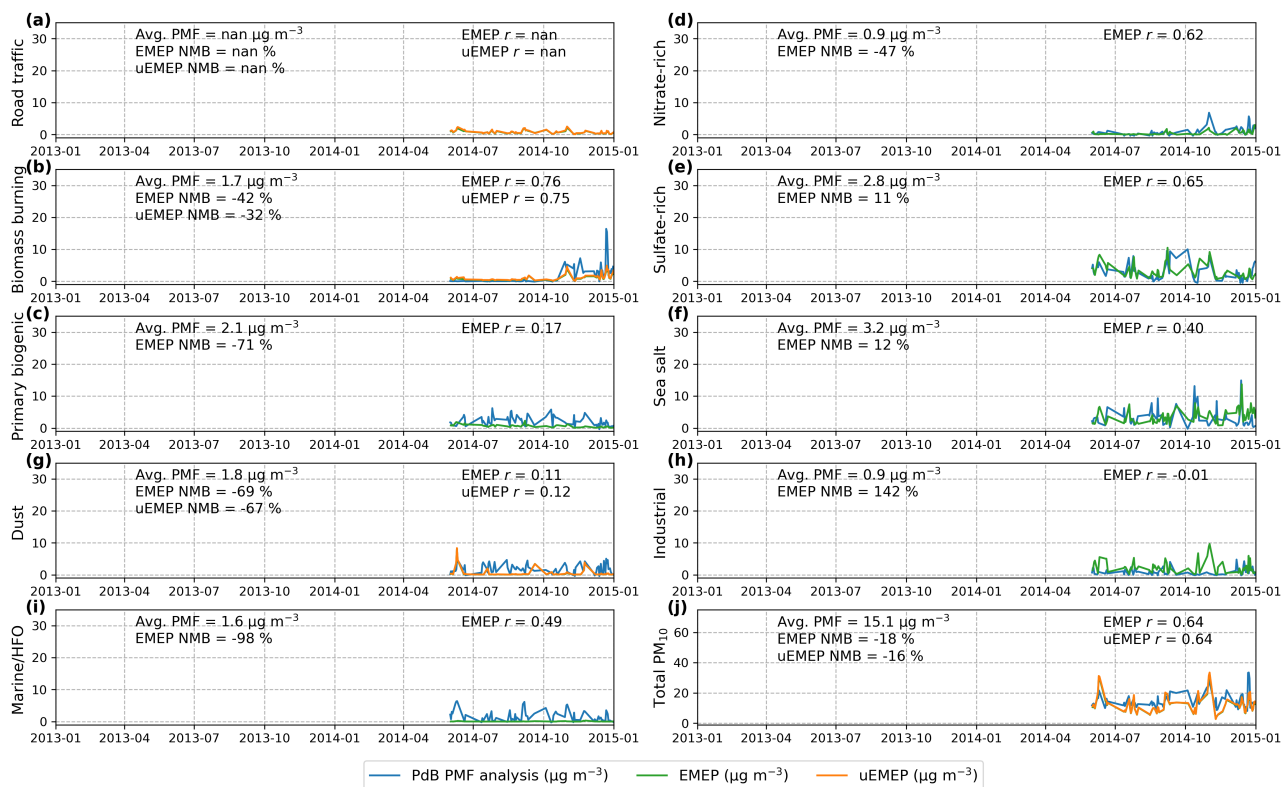


Figure S4. PMF derived and modelled surface concentrations at the PdB site. uEMEP results are shown only for those factors affected by downscaling. Note the difference in y-axis scaling in panel (h).

Table S4: Weber et al. (2019) station average PMF factor concentrations and EMEP and uEMEP model scores. Based on the stations and data availability shown in Fig. 1 of the main text. Note that here the TAL and Passy stations are included.

| Station | PMF factor | Avg. PMF ($\mu\text{g m}^{-3}$) | EMEP NMB | uEMEP NMB | EMEP r | uEMEP r |
|------------------|------------------------|-----------------------------------|----------|-----------|--------|---------|
| GRE-fr | Total PM ₁₀ | 17.8 | -16 % | 14 % | 0.63 | 0.60 |
| | Road traffic | 3.2 | -43 % | -3 % | 0.65 | 0.62 |
| | Biomass burning | 4.0 | 5 % | 98 % | 0.79 | 0.63 |
| | Dust | 2.8 | -59 % | -43 % | 0.20 | 0.21 |
| | Sulfate-rich | 3.0 | -9 % | | 0.56 | |
| | Nitrate-rich | 2.4 | 27 % | | 0.61 | |
| | Sea salt | 0.7 | -2 % | | 0.37 | |
| | Primary biogenic | 1.8 | -44 % | | 0.62 | |
| AIX | Total PM ₁₀ | 17.1 | -28 % | -12 % | 0.68 | 0.74 |
| | Road traffic | 2.2 | -38 % | 0 % | 0.29 | 0.47 |
| | Biomass burning | 3.6 | -39 % | 6 % | 0.84 | 0.79 |
| | Dust | 1.8 | -40 % | -24 % | 0.18 | 0.21 |
| | Sulfate-rich | 0.8 | 286 % | | 0.29 | |
| | Nitrate-rich | 1.5 | -17 % | | 0.69 | |
| | Sea salt | 1.4 | 16 % | | 0.54 | |
| | Primary biogenic | 1.7 | -36 % | | 0.23 | |
| | Industrial | 0.5 | 3 % | | 0.43 | |
| | Marine/HFO | 3.6 | -99 % | | 0.44 | |
| | LEN | Total PM ₁₀ | 18.8 | -10 % | -4 % | 0.81 |
| Road traffic | | 2.9 | -60 % | -49 % | 0.60 | 0.61 |
| Biomass burning | | 2.3 | 5 % | 34 % | 0.76 | 0.76 |
| Sulfate-rich | | 4.0 | -28 % | | 0.66 | |
| Nitrate-rich | | 5.8 | 3 % | | 0.79 | |
| Sea salt | | 3.1 | -10 % | | 0.67 | |
| Primary biogenic | | 0.6 | -13 % | | 0.39 | |
| REV | Total PM ₁₀ | 12.9 | -2 % | -3 % | 0.73 | 0.73 |
| | Road traffic | 1.9 | -70 % | -70 % | 0.76 | 0.76 |
| | Biomass burning | 1.5 | 7 % | -5 % | 0.74 | 0.71 |
| | Dust | 1.8 | -84 % | -84 % | 0.57 | 0.57 |
| | Sulfate-rich | 2.1 | 29 % | | 0.59 | |
| | Nitrate-rich | 3.5 | 26 % | | 0.70 | |

| | | | | | | |
|--------|------------------------|------|-------|-------|-------|------|
| | Sea salt | 1.5 | 34 % | | 0.60 | |
| | Primary biogenic | 0.7 | -2 % | | 0.54 | |
| CHAM | Total PM ₁₀ | 16.0 | -60 % | 37 % | 0.27 | 0.51 |
| | Road traffic | 0.9 | -55 % | 0 % | 0.30 | 0.35 |
| | Biomass burning | 7.3 | -84 % | 122 % | 0.59 | 0.56 |
| | Dust | 1.4 | -26 % | -14 % | 0.31 | 0.31 |
| | Sulfate-rich | 2.2 | -38 % | | 0.32 | |
| | Nitrate-rich | 1.2 | 26 % | | 0.17 | |
| | Sea salt | 1.1 | -70 % | | 0.35 | |
| | Primary biogenic | 1.8 | -85 % | | 0.61 | |
| Marnaz | Total PM ₁₀ | 14.5 | -8 % | -3 % | 0.59 | 0.53 |
| | Road traffic | 1.2 | -6 % | 21 % | 0.63 | 0.55 |
| | Biomass burning | 3.8 | 5 % | 14 % | 0.81 | 0.74 |
| | Dust | 2.5 | -64 % | -59 % | 0.27 | 0.28 |
| | Sulfate-rich | 2.6 | 6 % | | 0.43 | |
| | Nitrate-rich | 1.3 | 95 % | | 0.63 | |
| | Sea salt | 0.6 | -34 % | | 0.36 | |
| | Primary biogenic | 2.5 | -45 % | | 0.48 | |
| NGT | Total PM ₁₀ | 21.4 | -30 % | -26 % | 0.73 | 0.74 |
| | Road traffic | 4.3 | -76 % | -67 % | 0.64 | 0.64 |
| | Biomass burning | 3.3 | -39 % | -26 % | 0.85 | 0.81 |
| | Sulfate-rich | 3.5 | -27 % | | 0.61 | |
| | Nitrate-rich | 4.4 | 20 % | | 0.68 | |
| | Sea salt | 4.9 | -51 % | | 0.55 | |
| | Primary biogenic | 0.9 | -32 % | | 0.54 | |
| NIC | Total PM ₁₀ | 16.1 | -33 % | -10 % | 0.65 | 0.68 |
| | Road traffic | 1.7 | -24 % | 73 % | 0.43 | 0.59 |
| | Biomass burning | 2.1 | -35 % | 33 % | 0.84 | 0.88 |
| | Dust | 3.6 | -79 % | -63 % | 0.46 | 0.51 |
| | Sulfate-rich | 2.5 | 26 % | | 0.65 | |
| | Nitrate-rich | 0.7 | -23 % | | 0.41 | |
| | Sea salt | 2.2 | -18 % | | 0.37 | |
| | Primary biogenic | 1.4 | 6 % | | -0.21 | |
| | Marine/HFO | 1.8 | -97 % | | 0.33 | |
| NGT | Total PM ₁₀ | 21.4 | -30 % | -26 % | 0.73 | 0.74 |

| | | | | | | |
|---------|------------------------|------|-------|-------|-------|------|
| | Road traffic | 4.3 | -76 % | -67 % | 0.64 | 0.64 |
| | Biomass burning | 3.3 | -39 % | -26 % | 0.85 | 0.81 |
| | Sulfate-rich | 3.5 | -27 % | | 0.61 | |
| | Nitrate-rich | 4.4 | 20 % | | 0.68 | |
| | Sea salt | 4.9 | -51 % | | 0.55 | |
| | Primary biogenic | 0.9 | -32 % | | 0.54 | |
| PdB | Total PM ₁₀ | 15.1 | -18 % | -16 % | 0.64 | 0.64 |
| | Biomass burning | 1.7 | -42 % | -32 % | 0.76 | 0.75 |
| | Dust | 1.8 | -69 % | -67 % | 0.11 | 0.12 |
| | Sulfate-rich | 3.6 | -4 % | | 0.68 | |
| | Nitrate-rich | 0.9 | -47 % | | 0.62 | |
| | Sea salt | 2.4 | 35 % | | 0.48 | |
| | Primary biogenic | 2.1 | -71 % | | 0.17 | |
| | Industrial | 0.9 | 142 % | | -0.01 | |
| | Marine/HFO | 1.6 | -98 % | | 0.49 | |
| RBX | Total PM ₁₀ | 23.3 | -18 % | -5 % | 0.77 | 0.76 |
| | Road traffic | 2.4 | -36 % | 26 % | 0.28 | 0.39 |
| | Biomass burning | 2.3 | 29 % | 74 % | 0.75 | 0.75 |
| | Dust | 4.4 | -87 % | -75 % | 0.55 | 0.63 |
| | Sulfate-rich | 2.8 | 8 % | | 0.46 | |
| | Nitrate-rich | 6.5 | 2 % | | 0.71 | |
| | Sea salt | 3.7 | -24 % | | 0.53 | |
| | Primary biogenic | 1.1 | -59 % | | 0.41 | |
| STG-cle | Total PM ₁₀ | 17.0 | -7 % | 23 % | 0.77 | 0.73 |
| | Road traffic | 3.8 | -50 % | 4 % | 0.53 | 0.42 |
| | Biomass burning | 2.5 | 51 % | 150 % | 0.73 | 0.71 |
| | Dust | 1.8 | -72 % | -33 % | 0.20 | 0.24 |
| | Sulfate-rich | 3.9 | -22 % | | 0.61 | |
| | Nitrate-rich | 2.4 | 54 % | | 0.68 | |
| | Sea salt | 0.8 | -5 % | | 0.65 | |
| | Primary biogenic | 1.8 | -46 % | | 0.28 | |
| ROU | Total PM ₁₀ | 17.0 | -10 % | -4 % | 0.73 | 0.72 |
| | Road traffic | 5.3 | -79 % | -69 % | 0.57 | 0.58 |
| | Biomass burning | 1.0 | 117 % | 141 % | 0.64 | 0.58 |
| | Dust | 0.8 | -50 % | -27 % | 0.14 | 0.19 |

| | | | | | | |
|-------|------------------------|------|--------|-------|------|------|
| | Sulfate-rich | 4.3 | -37 % | | 0.62 | |
| | Nitrate-rich | 4.0 | 26 % | | 0.68 | |
| | Sea salt | 1.5 | 88 % | | 0.68 | |
| | Primary biogenic | 0.1 | 578 % | | 0.55 | |
| TAL | Total PM ₁₀ | 25.8 | -43 % | -36 % | 0.79 | 0.79 |
| | Road traffic | 1.3 | 13 % | 42 % | 0.60 | 0.55 |
| | Biomass burning | 6.5 | -49 % | -29 % | 0.68 | 0.67 |
| | Dust | 1.7 | -72 % | -64 % | 0.60 | 0.57 |
| | Sulfate-rich | 0.2 | 1097 % | | 0.64 | |
| | Nitrate-rich | 10.0 | -57 % | | 0.88 | |
| | Sea salt | 2.8 | -38 % | | 0.79 | |
| | Primary biogenic | 0.5 | 25 % | | 0.39 | |
| | Industrial | 1.8 | -78 % | | 0.57 | |
| | Marine/HFO | 1.1 | -96 % | | 0.65 | |
| Passy | Total PM ₁₀ | 28.3 | -71 % | -50 % | 0.45 | 0.58 |
| | Road traffic | 3.0 | -79 % | -58 % | 0.30 | 0.44 |
| | Biomass burning | 11.0 | -79 % | -35 % | 0.80 | 0.71 |
| | Dust | 5.4 | -91 % | -86 % | 0.19 | 0.21 |
| | Sulfate-rich | 3.6 | -51 % | | 0.33 | |
| | Nitrate-rich | 2.0 | -5 % | | 0.37 | |
| | Sea salt | 1.0 | -68 % | | 0.38 | |
| | Primary biogenic | 2.2 | -71 % | | 0.61 | |

Table S5. Model matching for the Daellenbach et al. (2020b, 2017) PMF factors. A discussion of the PMF factors as derived from measurement data is given in the main text.

| Modelled PMF factor | Assigned model outputs |
|---------------------|--|
| HOA | All modelled primary OA from GNFR F (road transport) vehicular exhaust, and all primary OA from offroad vehicles (GNFR I) and shipping (GNFR G). |
| SCOA | All modelled primary OA from GNFR F (road transport) tyre wear. |
| Brake wear metals | All modelled Cu and Fe from GNFR F (road transport) brake wear, with modelled brake wear assumed to be comprised of 70 % Cu and Fe. |
| BBOA | All primary OA from GNFR C (stationary combustion) and all primary OA from forest fires. |
| aSOA | All modelled aSOA. |
| bSOA | All modelled bSOA and PBAPs. |

Table S6: Daellenbach et al. (2020b, 2017) station average PMF factor concentrations and EMEP and uEMEP model scores. The OOA* factor is constructed as the sum of the aSOA and bSOA factors.

| Station | PMF factor | Avg. PMF ($\mu\text{g m}^{-3}$) | EMEP NMB | uEMEP NMB | EMEP r | uEMEP r |
|----------|-------------|-----------------------------------|----------|-----------|--------|---------|
| Bern | Vehic. wear | 1.4 | -85 % | -42 % | 0.65 | 0.58 |
| | Total OA | 6.1 | -7 % | 42 % | 0.47 | 0.42 |
| | HOA | 0.7 | -70 % | -34 % | 0.07 | 0.15 |
| | BBOA | 1.4 | 74 % | 240 % | 0.39 | 0.41 |
| | SCOA | 1.4 | -91 % | -68 % | 0.21 | 0.06 |
| | aSOA | 1.3 | -74 % | | 0.30 | |
| | bSOA | 1.2 | 101 % | | 0.76 | |
| | OOA* | 2.5 | 12 % | | 0.58 | |
| Basel | Total OA | 5.1 | 24 % | 99 % | 0.65 | 0.50 |
| | HOA | 0.8 | -67 % | -47 % | 0.43 | 0.42 |
| | BBOA | 1.2 | 173 % | 449 % | 0.66 | 0.55 |
| | SCOA | 0.6 | -83 % | -56 % | 0.21 | 0.13 |
| | aSOA | 1.1 | -66 % | | 0.10 | |
| | bSOA | 1.3 | 70 % | | 0.89 | |
| | OOA* | 2.4 | 6 % | | 0.63 | |
| Magadino | Vehic. wear | 0.3 | -62 % | -52 % | 0.82 | 0.19 |
| | Total OA | 7.2 | -49 % | -49 % | 0.15 | 0.12 |
| | HOA | 0.5 | -75 % | -74 % | 0.36 | 0.32 |
| | BBOA | 2.9 | -80 % | -80 % | 0.29 | 0.19 |
| | SCOA | 0.6 | -87 % | -84 % | 0.49 | 0.23 |
| | aSOA | 1.6 | -76 % | | -0.02 | |
| | bSOA | 1.6 | 56 % | | 0.82 | |
| | OOA* | 3.2 | -10 % | | 0.52 | |
| Payerne | Vehic. wear | 0.1 | 14 % | 85 % | 0.65 | 0.69 |
| | Total OA | 4.3 | -11 % | -6 % | 0.71 | 0.72 |
| | HOA | 0.5 | -77 % | -73 % | 0.43 | 0.42 |
| | BBOA | 0.9 | 12 % | 28 % | 0.58 | 0.59 |
| | SCOA | 0.4 | -89 % | -84 % | 0.18 | 0.15 |
| | aSOA | 1.1 | -71 % | | 0.24 | |
| | bSOA | 1.5 | 62 % | | 0.86 | |
| | OOA* | 2.5 | 7 % | | 0.67 | |

| | | | | | | |
|------------|-------------|----------|-------|-------|-------|-------|
| Zurich | Vehic. wear | 0.6 | -54 % | -13 % | 0.65 | 0.54 |
| | Total OA | 5.0 | 51 % | 126 % | 0.66 | 0.56 |
| | HOA | 0.7 | -54 % | -41 % | 0.40 | 0.36 |
| | BBOA | 1.1 | 268 % | 589 % | 0.57 | 0.50 |
| | SCOA | 0.8 | -78 % | -61 % | 0.57 | 0.47 |
| | aSOA | 1.1 | -63 % | | 0.28 | |
| | bSOA | 1.4 | 84 % | | 0.83 | |
| | OOA* | 2.5 | 21 % | | 0.65 | |
| Frauenland | Total OA | 5.3 | -19 % | 5 % | 0.63 | 0.71 |
| | HOA | 0.7 | -78 % | -58 % | 0.26 | 0.24 |
| | BBOA | 1.1 | 15 % | 100 % | 0.56 | 0.60 |
| | SCOA | 1.1 | -93 % | -78 % | 0.44 | 0.33 |
| | aSOA | 1.0 | -65 % | | 0.09 | |
| | bSOA | 1.4 | 78 % | | 0.87 | |
| | OOA* | 2.4 | 17 % | | 0.59 | |
| | Gallen | Total OA | 4.4 | -11 % | 47 % | 0.69 |
| HOA | | 0.5 | -72 % | -35 % | 0.36 | 0.37 |
| BBOA | | 0.8 | 52 % | 329 % | 0.52 | 0.36 |
| SCOA | | 0.9 | -93 % | -65 % | 0.46 | 0.35 |
| aSOA | | 0.9 | -65 % | | 0.16 | |
| bSOA | | 1.3 | 67 % | | 0.82 | |
| OOA* | | 2.2 | 13 % | | 0.59 | |
| Vaduz | | Total OA | 5.0 | -25 % | -3 % | 0.44 |
| | HOA | 0.6 | -86 % | -53 % | 0.20 | 0.14 |
| | BBOA | 1.1 | -38 % | 27 % | 0.38 | 0.38 |
| | SCOA | 1.0 | -96 % | -77 % | 0.56 | 0.35 |
| | aSOA | 1.0 | -74 % | | 0.19 | |
| | bSOA | 1.3 | 107 % | | 0.70 | |
| | OOA* | 2.3 | 28 % | | 0.46 | |
| | Vittore | Total OA | 10.5 | -72 % | -71 % | -0.15 |
| HOA | | 0.4 | -85 % | -78 % | 0.11 | 0.16 |
| BBOA | | 6.4 | -94 % | -94 % | 0.07 | -0.08 |
| SCOA | | 0.7 | -96 % | -90 % | -0.13 | -0.05 |
| aSOA | | 1.5 | -78 % | | -0.04 | |
| | | | | | | |

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|------|-----|-------|------|
| bSOA | 1.5 | 43 % | 0.84 |
| OOA* | 3.0 | -17 % | 0.41 |

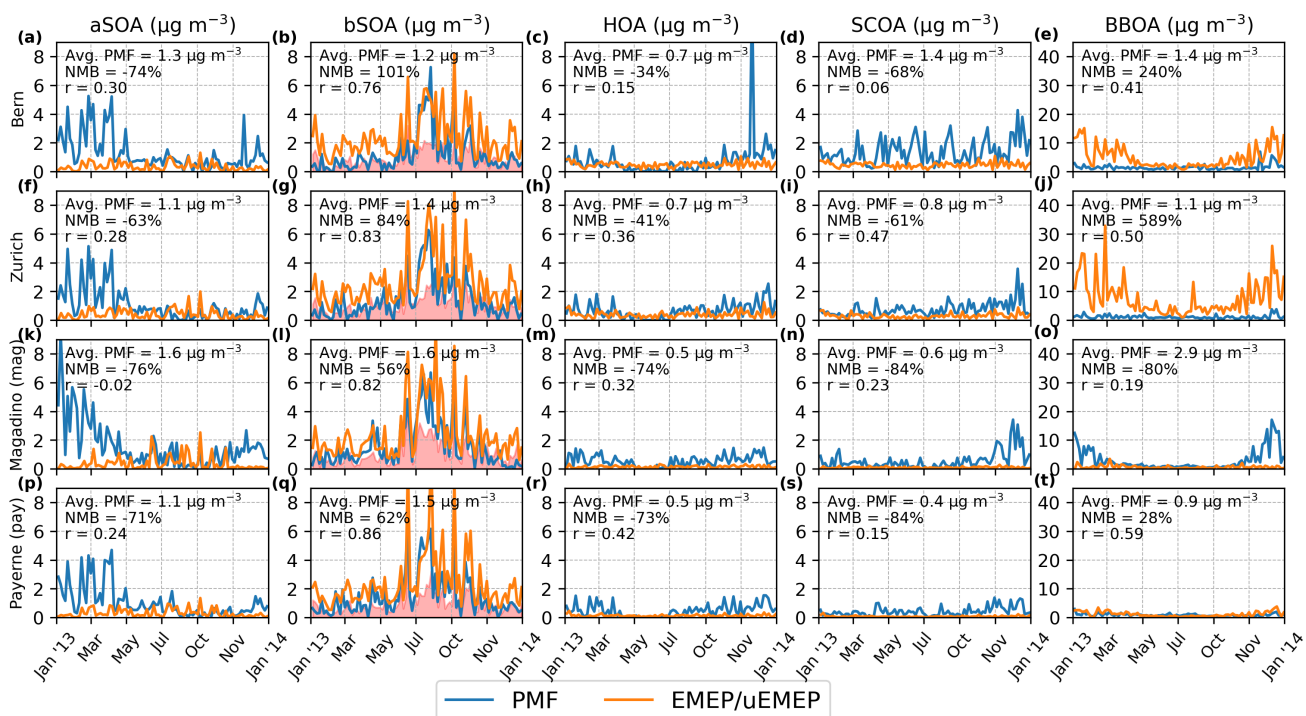


Figure S5. Comparison of PMF derived OA fractions from Daellenbach et al. (2017) and the corresponding EMEP/uEMEP modelling results. Note the difference in y-axis scaling for the right-most panels. Shading in the panels showing bSOA results indicate the modelled contribution of PBAPs to this factor.

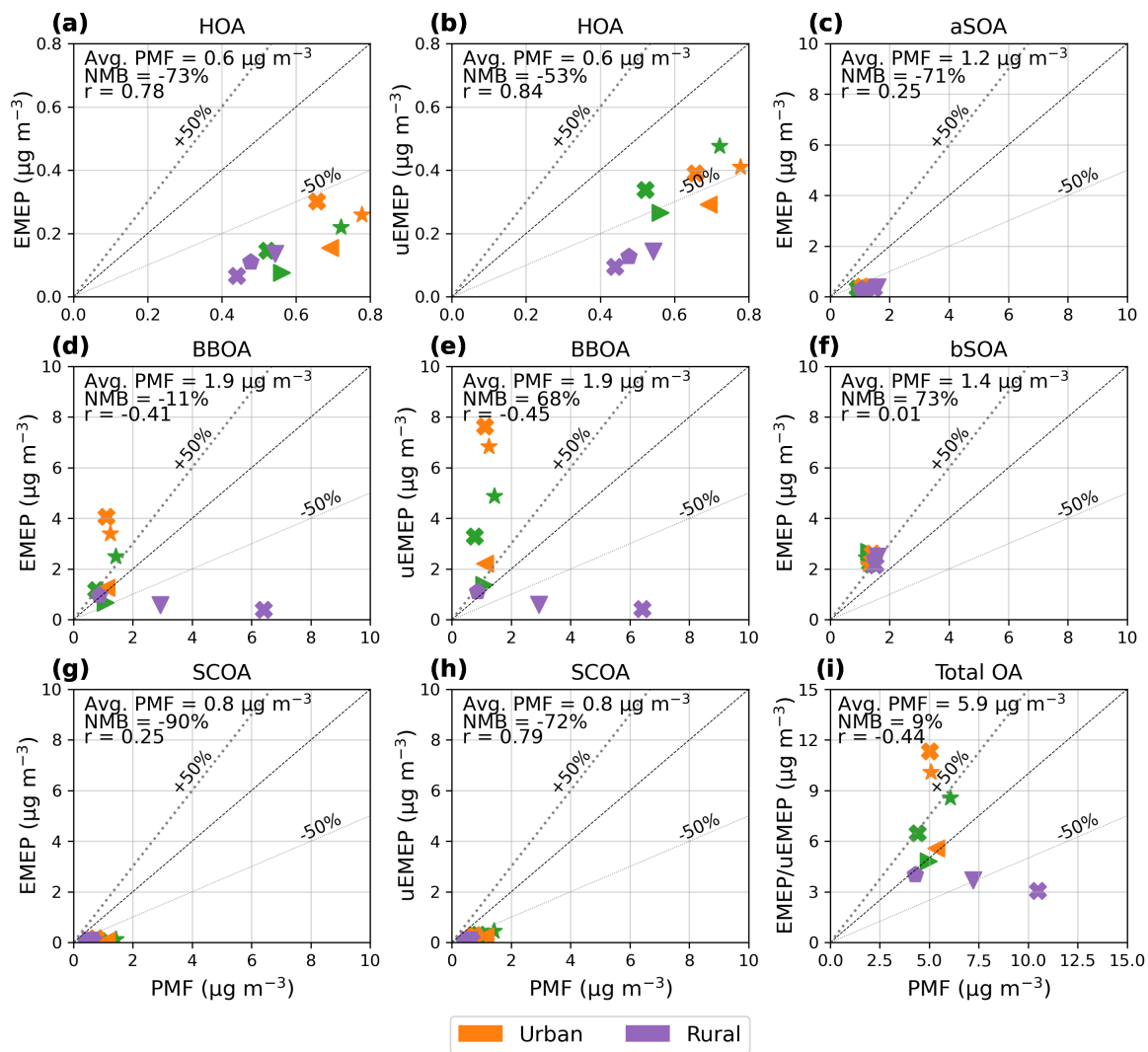


Figure S6. Average observed and simulated OA PMF factor concentrations at the Daellenbach et al. (2017) sites. Markers correspond to those shown in Fig. 1 of the main text. Note the differences in axis scaling for panel (i).

Table S7. Model matching for the Chen et al. (2022) PMF factors. A discussion of the PMF factors as derived from measurement data is given in the main text.

| Modelled PMF factor | Assigned model outputs |
|---------------------|---|
| HOA | All modelled fine-mode primary OA from GNFR F (road transport) vehicular exhaust, and all fine-mode primary OA from off-road vehicles (GNFR I) and shipping (GNFR G). |
| BBOA | All fine-mode primary OA from GNFR C (stationary combustion) and all fine-mode primary OA from forest fires. |
| OOA | All modelled aSOA and bSOA. |

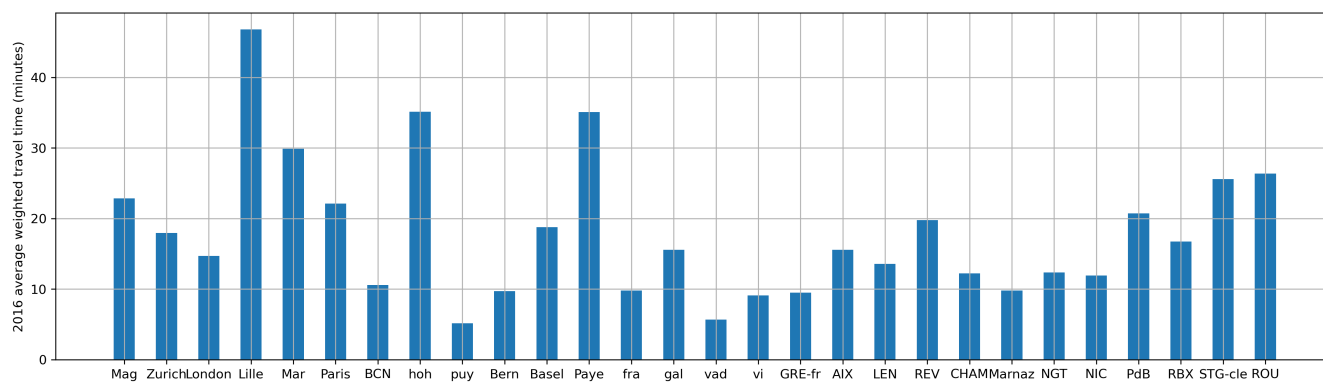


Figure S7. Average weighted travel times of the downscaled uEMEP PM₁₀ at the sites discussed in the main text.

Table S8. Chen *et al.* (2022) station average PMF factor concentrations and EMEP and uEMEP model scores based on daily averaged data.

| Station | PMF factor | Avg. obs ($\mu\text{g m}^{-3}$) | EMEP NMB | uEMEP NMB | EMEP r | uEMEP r |
|------------------|------------|-----------------------------------|----------|-----------|--------|---------|
| Magadino | HOA | 0.5 | -79 % | -77 % | 0.54 | 0.48 |
| | BBOA | 0.8 | -40 % | -38 % | 0.51 | 0.44 |
| | OOA | 4.3 | -59 % | -59 % | 0.73 | 0.73 |
| | Total OA | 5.7 | -58 % | -57 % | 0.70 | 0.69 |
| Zurich | HOA | 0.2 | 52 % | 84 % | 0.41 | 0.44 |
| | BBOA | 0.5 | 959 % | 1697 % | 0.26 | 0.27 |
| | OOA | 1.0 | 84 % | 84 % | 0.82 | 0.82 |
| | Total OA | 1.7 | 325 % | 536 % | 0.42 | 0.38 |
| London | HOA | 0.5 | -44 % | -41 % | 0.75 | 0.73 |
| | BBOA | 1.2 | -43 % | -44 % | 0.68 | 0.68 |
| | OOA | 2.2 | -58 % | -58 % | 0.46 | 0.46 |
| | Total OA | 3.8 | -52 % | -51 % | 0.77 | 0.77 |
| Lille | HOA | 0.6 | -61 % | -50 % | 0.68 | 0.66 |
| | BBOA | 0.7 | 279 % | 298 % | 0.78 | 0.80 |
| | OOA | 3.5 | -66 % | -66 % | 0.60 | 0.60 |
| | Total OA | 4.8 | -15 % | -11 % | 0.86 | 0.86 |
| Marseille | HOA | 0.4 | -76 % | -70 % | 0.48 | 0.50 |
| | BBOA | 1.2 | 53 % | 133 % | 0.78 | 0.79 |
| | OOA | 3.1 | -43 % | -43 % | 0.37 | 0.37 |
| | Total OA | 4.7 | -23 % | -3 % | 0.70 | 0.76 |
| Paris | HOA | 0.5 | -55 % | -54 % | 0.67 | 0.67 |
| | BBOA | 0.7 | 165 % | 157 % | 0.78 | 0.79 |
| | OOA | 3.1 | -61 % | -61 % | 0.58 | 0.58 |
| | Total OA | 4.3 | -22 % | -24 % | 0.88 | 0.88 |
| Barcelona | HOA | 0.4 | -21 % | -60 % | 0.46 | 0.39 |
| | BBOA | 0.7 | 455 % | 417 % | 0.63 | 0.58 |
| | OOA | 2.9 | -11 % | -11 % | 0.81 | 0.81 |
| | Total OA | 4.0 | 65 % | 54 % | 0.67 | 0.67 |
| Hohenpeissenberg | HOA | 0.1 | -47 % | -43 % | 0.26 | 0.25 |
| | BBOA | 0.1 | 586 % | 651 % | 0.16 | 0.16 |
| | OOA | 2.8 | -30 % | -30 % | 0.69 | 0.69 |
| | Total OA | 3.0 | -11 % | -9 % | 0.75 | 0.74 |
| Puy de Dome | HOA | 0.5 | -91 % | -92 % | 0.45 | 0.44 |
| | OOA | 3.9 | -60 % | -60 % | 0.62 | 0.62 |
| | Total OA | 4.5 | -47 % | -48 % | 0.59 | 0.59 |

Table S9. Chen *et al.* (2022) station average PMF factor concentrations and EMEP and uEMEP model scores based on hourly averaged data.

| Station | PMF factor | Avg. obs ($\mu\text{g m}^{-3}$) | EMEP NMB | uEMEP NMB | uEMEP r | EMEP r |
|------------------|------------|-----------------------------------|----------|-----------|---------|--------|
| Magadino | HOA | 0.5 | -79% | -77% | 0.29 | 0.18 |
| | BBOA | 0.8 | -40% | -37% | 0.40 | 0.28 |
| | OOA | 4.3 | -59% | -59% | 0.61 | 0.61 |
| | Total OA | 5.6 | -58% | -57% | 0.58 | 0.56 |
| Zurich | HOA | 0.2 | 53% | 86% | 0.24 | 0.27 |
| | BBOA | 0.5 | 986% | 1736% | 0.24 | 0.27 |
| | OOA | 1.0 | 84% | 84% | 0.64 | 0.64 |
| | Total OA | 1.7 | 333% | 547% | 0.33 | 0.29 |
| London | HOA | 0.5 | -44% | -40% | 0.48 | 0.45 |
| | BBOA | 1.1 | -43% | -44% | 0.63 | 0.62 |
| | OOA | 2.1 | -58% | -58% | 0.44 | 0.44 |
| | Total OA | 3.7 | -51% | -51% | 0.71 | 0.70 |
| Lille | HOA | 0.6 | -61% | -50% | 0.42 | 0.36 |
| | BBOA | 0.7 | 276% | 295% | 0.67 | 0.59 |
| | OOA | 3.5 | -66% | -66% | 0.53 | 0.53 |
| | Total OA | 4.8 | -15% | -11% | 0.74 | 0.67 |
| Marseille | HOA | 0.4 | -75% | -69% | 0.34 | 0.36 |
| | BBOA | 1.1 | 55% | 133% | 0.64 | 0.56 |
| | OOA | 3.1 | -43% | -43% | 0.36 | 0.36 |
| | Total OA | 4.6 | -22% | -3% | 0.62 | 0.59 |
| Paris | HOA | 0.4 | -55% | -54% | 0.50 | 0.50 |
| | BBOA | 0.7 | 163% | 155% | 0.67 | 0.68 |
| | OOA | 3.1 | -61% | -61% | 0.53 | 0.53 |
| | Total OA | 4.2 | -23% | -24% | 0.74 | 0.75 |
| Barcelona | HOA | 0.4 | -18% | -60% | 0.41 | 0.28 |
| | BBOA | 0.6 | 458% | 419% | 0.44 | 0.36 |
| | OOA | 2.9 | -11% | -11% | 0.70 | 0.70 |
| | Total OA | 4.0 | 65% | 54% | 0.46 | 0.42 |
| Hohenpeissenberg | HOA | 0.1 | -47% | -43% | 0.18 | 0.17 |
| | BBOA | 0.1 | 587% | 652% | 0.14 | 0.14 |
| | OOA | 2.8 | -30% | -30% | 0.60 | 0.60 |
| | Total OA | 3.0 | -11% | -9% | 0.63 | 0.63 |
| Puy de Dome | HOA | 0.6 | -91% | -92% | 0.30 | 0.30 |
| | OOA | 4.4 | -61% | -61% | 0.54 | 0.54 |
| | Total OA | 4.9 | -49% | -50% | 0.48 | 0.49 |

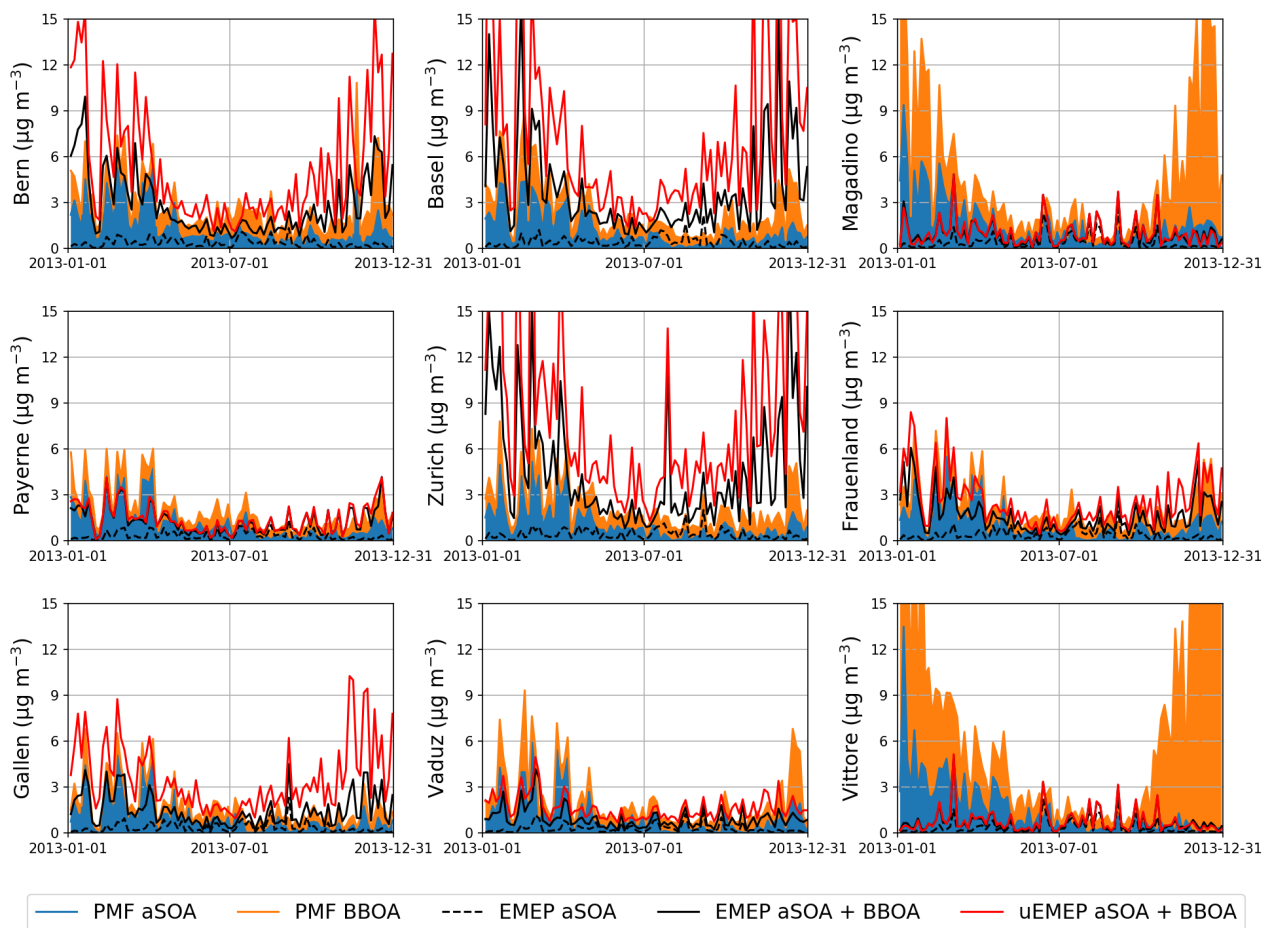


Figure S8. Comparison of PMF derived aSOA and BBOA contributions and the EMEP and uEMEP modelled contributions at the Daellenbach et al. (2017) sites. The PMF contributions are shown as cumulative shaded areas.

Table S10. Statistics based on daily mean data from all PMF datasets, comparing the model configurations where a volatile CPOA ratio of 0.5 is assumed (indicated by EMEP* and uEMEP*). The OOA* factor is constructed as the sum of the aSOA and bSOA factors.

| Dataset | PMF factor | Stations | Avg. PMF ($\mu\text{g m}^{-3}$) | EMEP* NMB | uEMEP* NMB | EMEP* r | uEMEP* r | EMEP* r_t | uEMEP* r_t |
|-------------|------------------------|----------|--------------------------------------|--------------|---------------|------------|-------------|----------------|-----------------|
| Weber | Total PM ₁₀ | 12 | 17.5 | -19 % | -6 % | 0.62 | 0.74 | 0.67 | 0.68 |
| | Road traffic | 11 | 2.9 | -57 % | -30 % | 0.37 | 0.23 | 0.52 | 0.54 |
| | Biomass burning | 12 | 2.9 | -13 % | 29 % | 0.04 | 0.87 | 0.76 | 0.74 |
| | Dust | 10 | 2.3 | -69 % | -56 % | 0.14 | 0.42 | 0.30 | 0.33 |
| | Sulfate-rich | 12 | 3.1 | -10 % | -10 % | 0.13 | 0.13 | 0.54 | 0.54 |
| | Nitrate-rich | 12 | 3.3 | 17 % | 17 % | 0.96 | 0.96 | 0.61 | 0.61 |
| | Sea salt | 12 | 2.0 | -11 % | -11 % | 0.70 | 0.70 | 0.51 | 0.51 |
| | Primary biogenic | 12 | 1.3 | -40 % | -40 % | 0.43 | 0.43 | 0.38 | 0.38 |
| | Industrial | 2 | 0.7 | 104 % | 104 % | 1.00 | 1.00 | 0.21 | 0.21 |
| | Marine/HFO | 3 | 2.2 | -98 % | -98 % | 0.35 | 0.07 | 0.42 | 0.49 |
| Daellenbach | Total OA | 9 | 5.9 | -21 % | -3 % | -0.40 | -0.44 | 0.47 | 0.45 |
| | HOA | 9 | 0.6 | -73 % | -53 % | 0.78 | 0.84 | 0.29 | 0.29 |
| | BBOA | 9 | 1.9 | -55 % | -15 % | -0.41 | -0.45 | 0.44 | 0.39 |
| | SCOA | 9 | 0.8 | -90 % | -72 % | 0.25 | 0.79 | 0.33 | 0.22 |
| | aSOA | 9 | 1.2 | -1 % | -1 % | -0.22 | -0.22 | 0.44 | 0.44 |
| | bSOA | 9 | 1.4 | 73 % | 73 % | 0.01 | 0.01 | 0.82 | 0.82 |
| | OOA* | 9 | 2.6 | 39 % | 39 % | -0.33 | -0.33 | 0.57 | 0.57 |
| | Vehic. wear | 4 | 0.5 | -68 % | -31 % | 0.61 | 0.96 | 0.69 | 0.50 |
| Chen | Total OA | 8 | 4.2 | -25 % | -24 % | -0.03 | 0.02 | 0.74 | 0.75 |
| | HOA | 8 | 0.4 | -58 % | -59 % | 0.28 | 0.43 | 0.54 | 0.52 |
| | BBOA | 7 | 0.8 | -2 % | 5 % | -0.01 | 0.13 | 0.60 | 0.60 |
| | OOA | 8 | 3.0 | -26 % | -26 % | -0.01 | -0.01 | 0.73 | 0.73 |