S1 Summary of instruments and their availability during the campaign

Parameter	Instrument	Frequency	Detection limit
SAFIRE ATR-42 research aircraft			
Non-refractory Aerosol Particle Composition	mAMS (Aerodyne)	Measured at 1 s (FastMS mode), final data averaged over 15 s	Org: 0.08 µg m^{-3} SO_4: 0.007 µg m^{-3} NO_3: 0.008 µg m^{-3} Chl: 0.008 µg m^{-3} NH_4: 0.034 µg m^{-3}
Refractory Black Carbon (rBC)	SP2 (DMT)	1 s	< 10 ng m^{-3}
Aerosol extinction and scattering coefficient at 450 nm and 630 nm	A2S2 (dual CAPS-PM_{SSA}, Aerodyne)	Measured at 1 s, final data averaged over 10 s	10s integration: 0.69 Mm^{-1} for $\boldsymbol{\sigma_{ext,450\ nm}}$ 0.45 Mm^{-1} for $\boldsymbol{\sigma_{scat,450\ nm}}$ 0.21 Mm^{-1} for $\boldsymbol{\sigma_{ext,630\ nm}}$ 0.12 Mm^{-1} for $\boldsymbol{\sigma_{scat,630\ nm}}$
Particle Size Distribution (~100 - 1000 nm)	UHSAS (DMT)	1 s	-
Particle Size Distribution (~1 – 10 µm)	Sky-OPC (GRIMM)	6 s	-
NO_2	TD-LIF for most of the flights and CAPS-NO_2 (T500U, Teledyne) for Flight A036	1 s for TD-LIF 1 min for CAPS-NO_2	9.8 ppt for TD-LIF < 40 ppt for CAPS-NO_2
CO	Gas analyser (G2401-m, PICARO)	1 s	15 ppb
O_3	Ozone analyser (Model 49i, Thermo Scientific)	5 s	0.5 ppb
PRG ground site			
Non-refractory Aerosol Particle Composition	Time-of-Flight Aerosol Chemical Speciation Monitor (ToF-ACSM, Aerodyne)	~6 min	Org: 0.31 µg m^{-3} SO_4: 0.04 µg m^{-3} NO_3: 0.09 µg m^{-3} Chl: 0.06 µg m^{-3}

			NH_4: 0.19 µg m^{-3}
Aerosol absorption coefficient at 370, 470, 520, 590, 660, and 880 and 950 nm	Aethalometer (AE33, Magee Sci.)	1 min	-
Aerosol scattering coefficient at 450, 550, and 700 nm	Nephelometer (Ecotech Aurora 4000)	1 min	< 0.3 Mm^{-1} for all wavelengths
Particle Size Distribution (~0.02 – ~1 µm)	SMPS (DMA, TSI model 3081 and CPC, TSI model 3775)	~2.5 min	-
NO_2	AC32M monitor (Environment SA)	1 s	0.4 ppb
O_3	41M monitor (Environment SA)	1 s	1 ppb
CO	AIRPARIF network in central Paris	1 h	-

Table S1: Selected airborne plume sampling periods and their corresponding PRG ground-based measurement windows. The estimated average [minimum, maximum] plume travel time is also presented.

Instruments		A027(a)	A027(b)	A028	A031	A032	A033	A034	A036
SAFIRE aircraft									
mAMS									
SP2									
A2S2	**450 nm**								
	630 nm								
UHSAS									
Sky-GRIMM									
TD-LIF									CAPS-NO_2
O_3 gas analyser									
CO gas analyser									
PRG site									
ACSM									
NEPH									
AE33									
SMPS									
AC32M NO_2 monitor									
41M O_3 monitor									
AIRPARIF network CO									

Table S2: Availability of the instruments used in this study. Green indicates that the instrument was available for the entire period, red indicates that it was unavailable, and yellow indicates partial availability. TD-LIF was replaced by CAPS-NO2 for Flight A036.

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S3 Selected Paris urban plume period and estimated plume transport time

	Airborne measured plume periods (ATR 42)	Corresponding ground-based measurement windows (PRG)	Estimated average plume travel time [minimum, maximum]
A027(a) 21/06/2022	11:15:00 - 11:26:00 UTC	07:00:00 UTC - 10:00:00 UTC	2.4h [1.6h, 3.7h]
A027(b) (plume 1) 22/06/2022	13:12:00 - 13:21:00 UTC	09:00:00 - 12:00:00 UTC	2.9h [2.1h, 3.7h]
A027(b) (plume 2) 22/06/2022	13:24:00 - 13:29:30 UTC		3.0h [2.4h, 3.4h]
A028 (plume 1) 23/06/2022	11:14:00 - 11:18:00 UTC	06:00:00 - 09:00:00 UTC	3.1h [2.7h, 3.3h]
A028 (plume 2) 23/06/2022	11:23:00 - 11:28:00 UTC		3.6h [3.2h, 3.9h]
A031 (plume 1) 28/06/2022	10:13:00 - 10:17:30 UTC	04:00:00 - 07:00:00 UTC	4.9h [4.0h, 5.8h]
A031 (plume 2) 28/06/2022	10:23:00 - 10:28:00 UTC		5.4h [4.5h, 6.4h]
A032 (plume 1) 29/06/2022	11:13:30 - 11:17:00 UTC	06:00:00 - 09:00:00 UTC	2.9h [2.5h, 3.3h]
A032 (plume 2) 29/06/2022	11:23:00 - 11:27:00 UTC		3.9h [3.2h, 4.4h]
A033 (plume 1) 01/07/2022	11:24:30 - 11:30:00 UTC	06:00:00 - 09:00:00 UTC	2.9h [2.7h, 3.0h]
A033 (plume 2) 01/07/2022	11:42:00 - 11:46:00 UTC		3.5h [3.3h, 3.7h]
A034 (plume 1) 02/07/2022	11:08:00 - 11:14:00 UTC	06:00:00 - 09:00:00 UTC	4.1h [3.4h, 5.0h]
A034 (plume 2) 02/07/2022	11:30:00 - 11:34:00 UTC		5.9h [5.5h, 6.5h]
A036 (plume 1) 05/07/2022	13:31:00 - 13:36:00 UTC	08:00:00 - 11:00:00 UTC	4.2h [4.0h, 4.3h]
A036 (plume 2) 05/07/2022	13:41:00 - 13:48:00 UTC		3.4h [3.0h, 3.7h]

Table S3: Selected airborne plume sampling periods and their corresponding PRG ground-based measurement windows. The estimated average [minimum, maximum] plume travel time is also presented.

S4 Comparison of aerosol volume derived from UHSAS measurements with that estimated by mAMS

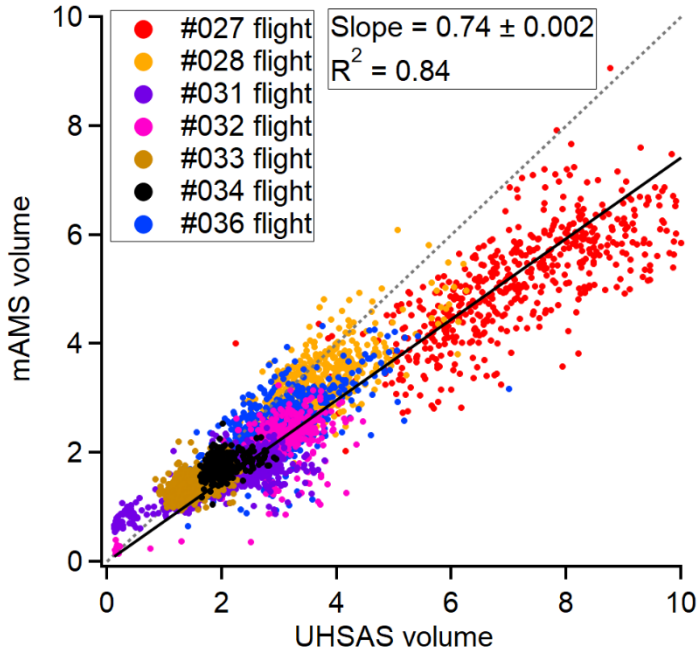

Figure S1: Comparison between the mAMS estimated aerosol volume and the UHSAS derived volume. The total mass concentrations measured from the mAMS were converted to total volume concentrations, using densities of 1.27 g cm^{-3} for organics, 1.77 g cm^{-3} for inorganics.

S5 Average airborne measured PM_1 components mass concentrations for each flight

	Org ($\mu g\ m^{-3}$)	**SO_4** ($\mu g\ m^{-3}$)	**NO_3** ($\mu g\ m^{-3}$)	**Chl** ($\mu g\ m^{-3}$)	**NH_4** ($\mu g\ m^{-3}$)	**rBC** ($\mu g\ m^{-3}$)
A027(a) (Plume) 22/06/2022	6.05 ± 0.43	1.86 ± 0.15	0.82 ± 0.17	0.13 ± 0.04	0.83 ± 0.08	N/A
A027(a) (Background) 22/06/2022	5.94 ± 0.74	1.85 ± 0.15	0.69 ± 0.15	0.15 ± 0.04	0.79 ± 0.07	N/A
A027(b) (Plume) 22/06/2022	6.53 ± 0.78	1.65 ± 0.20	0.95 ± 0.31	0.15 ± 0.03	0.79 ± 0.13	N/A
A027(b) (Background) 22/06/2022	6.37 ± 0.73	1.79 ± 0.16	0.65 ± 0.20	0.15 ± 0.03	0.76 ± 0.10	N/A
A028 (Plume) 23/06/2022	3.23 ± 0.39	0.76 ± 0.10	0.27 ± 0.10	0.09 ± 0.08	0.44 ± 0.06	0.18 ± 0.07
A028 (Background) 23/06/2022	2.80 ± 0.42	0.84 ± 0.14	0.26 ± 0.08	0.08 ± 0.08	0.46 ± 0.07	0.10 ± 0.05
A031 (Plume) 28/06/2022	1.47 ± 0.20	0.42 ± 0.05	0.22 ± 0.08	0.08 ± 0.02	0.23 ± 0.02	N/A
A031 (Background) 28/06/2022	1.34 ± 0.19	0.41 ± 0.03	0.18 ± 0.05	0.07 ± 0.02	0.21 ± 0.02	N/A
A032 (Plume) 29/06/2022	2.14 ± 0.24	0.43 ± 0.04	0.24 ± 0.07	0.09 ± 0.03	0.23 ± 0.02	0.18 ± 0.24
A032 (Background) 29/06/2022	1.86 ± 0.39	0.40 ± 0.04	0.15 ± 0.06	0.09 ± 0.03	0.20 ± 0.02	0.09 ± 0.16
A033 (Plume) 01/07/2022	1.23 ± 0.22	0.32 ± 0.04	0.25 ± 0.08	0.07 ± 0.02	0.19 ± 0.03	0.12 ± 0.05
A033 (Background) 01/07/2022	1.21 ± 0.18	0.28 ± 0.04	0.18 ± 0.05	0.07 ± 0.02	0.16 ± 0.02	0.04 ± 0.04
A034 (Plume) 02/07/2022	1.72 ± 0.18	0.35 ± 0.04	0.20 ± 0.05	0.08 ± 0.02	0.19 ± 0.02	N/A
A034 (Background) 02/07/2022	1.54 ± 0.17	0.33 ± 0.04	0.19 ± 0.06	0.08 ± 0.02	0.18 ± 0.02	N/A
A036 (Plume) 05/07/2022	3.15 ± 0.50	0.58 ± 0.06	0.30 ± 0.12	0.15 ± 0.04	0.31 ± 0.04	N/A
A036 (Background) 05/07/2022	1.91 ± 0.38	0.59 ± 0.10	0.15 ± 0.09	0.13 ± 0.04	0.26 ± 0.05	N/A

Table S4: Average airborne measured PM_1 components mass concentrations (reported as mean ± standard deviation) for each flight

S6 Average airborne measured optical parameters for each flight

	450 nm			630 nm		
	σ_{ext} (Mm^{-1})	σ_{scat} (Mm^{-1})	SSA	σ_{ext} (Mm^{-1})	σ_{scat} (Mm^{-1})	SSA
A027(a) (Plume) 22/06/2022	67.02 ± 8.08	58.09 ± 6.90	0.87 ± 0.04	39.40 ± 5.21	33.49 ± 4.42	0.85 ± 0.04
A027(a) (Background) 22/06/2022	55.27 ± 6.36	49.64 ± 4.56	0.90 ± 0.07	32.23 ± 4.42	27.77 ± 3.51	0.86 ± 0.03
A027(b) (Plume) 22/06/2022	81.62 ± 13.73	68.51 ± 12.08	0.84 ± 0.06	N/A	N/A	N/A
A027(b) (Background) 22/06/2022	58.33 ± 5.63	53.67 ± 7.55	0.92 ± 0.06	N/A	N/A	N/A
A028 (Plume) 23/06/2022	N/A	N/A	N/A	21.35 ± 3.22	17.49 ± 2.33	0.83 ± 0.10
A028 (Background) 23/06/2022	N/A	N/A	N/A	15.82 ± 2.03	13.44 ± 1.73	0.85 ± 0.05
A031 (Plume) 28/06/2022	13.14 ± 2.23	11.70 ± 1.72	0.90 ± 0.08	9.23 ± 2.20	7.79 ± 1.56	0.86 ± 0.08
A031 (Background) 28/06/2022	8.90 ± 1.20	8.31 ± 0.83	0.94 ± 0.05	6.05 ± 1.55	5.54 ± 1.60	0.91 ± 0.10
A032 (Plume) 29/06/2022	22.03 ± 3.69	17.77 ± 2.03	0.82 ± 0.10	14.42 ± 2.27	12.24 ± 2.10	0.85 ± 0.06
A032 (Background) 29/06/2022	15.43 ± 3.30	12.95 ± 2.67	0.85 ± 0.09	9.29 ± 2.74	8.43 ± 2.42	0.91 ± 0.08
A033 (Plume) 01/07/2022	8.04 ± 1.84	7.06 ± 1.42	0.89 ± 0.10	6.26 ± 2.55	5.61 ± 2.15	0.91 ± 0.10
A033 (Background) 01/07/2022	5.57 ± 1.06	5.13 ± 0.96	0.93 ± 0.10	4.87 ± 2.49	4.47 ± 2.29	0.93 ± 0.10
A034 (Plume) 02/07/2022	11.09 ± 1.09	9.62 ± 1.16	0.87 ± 0.10	6.44 ± 1.80	5.41 ± 1.60	0.85 ± 0.14
A034 (Background) 02/07/2022	7.04 ± 1.95	6.44 ± 1.36	0.93 ± 0.10	4.78 ± 2.27	4.38 ± 2.14	0.93 ± 0.11
A036 (Plume) 05/07/2022	25.69 ± 4.47	21.40 ± 3.19	0.84 ± 0.08	15.94 ± 3.08	13.37 ± 2.39	0.84 ± 0.05
A036 (Background) 05/07/2022	15.85 ± 2.37	13.49 ± 1.74	0.86 ± 0.07	8.79 ± 1.66	7.49 ± 1.19	0.86 ± 0.08

Table S5: Average airborne measured optical parameters (reported as mean ± standard deviation) for each flight

S7 Average airborne measured metrological parameters for each flight

	Ambient T (℃)	**Ambient RH** (%)	**RH measured at inlet** (%)	**Horizontal wind direction** (degree)	**Horizontal wind speed** (m s^{-1})
A027(a) 22/06/2022	19.1 ± 0.5	68 ± 7	37 ± 2	68.97 ± 17.51	3.96 ± 0.93
A027(b) 22/06/2022	18.1 ± 0.5	74 ± 7	27 ± 2	81.39 ± 13.82	5.39 ± 0.97
A028 23/06/2022	19.6 ± 0.5	67 ± 3	30 ± 1	193.25 ± 12.67	5.56 ± 1.00
A031 28/06/2022	16.1 ± 0.2	56 ± 2	22 ± 1	188.95 ± 15.39	5.68 ± 1.27
A032 29/06/2022	18.5 ± 0.7	47 ± 2	20 ± 1	209.05 ± 15.84	5.86 ± 1.33
A033 01/07/2022	14.0 ± 0.3	57 ± 2	22 ± 1	253.68 ± 15.66	4.96 ± 1.07
A034 02/07/2022	17.5 ± 0.6	46 ± 2	21 ± 1	192.44 ± 23.18	4.01 ± 1.61
A036 05/07/2022	19.8 ± 0.3	45 ± 3	17 ± 1	309.55 ± 67.37	4.06 ± 1.10

35 **Table S6: Average airborne measured metrological parameters (reported as mean ± standard deviation) for each flight**

S8 Average PM_1 component mass fraction at PRG and downwind urban plume

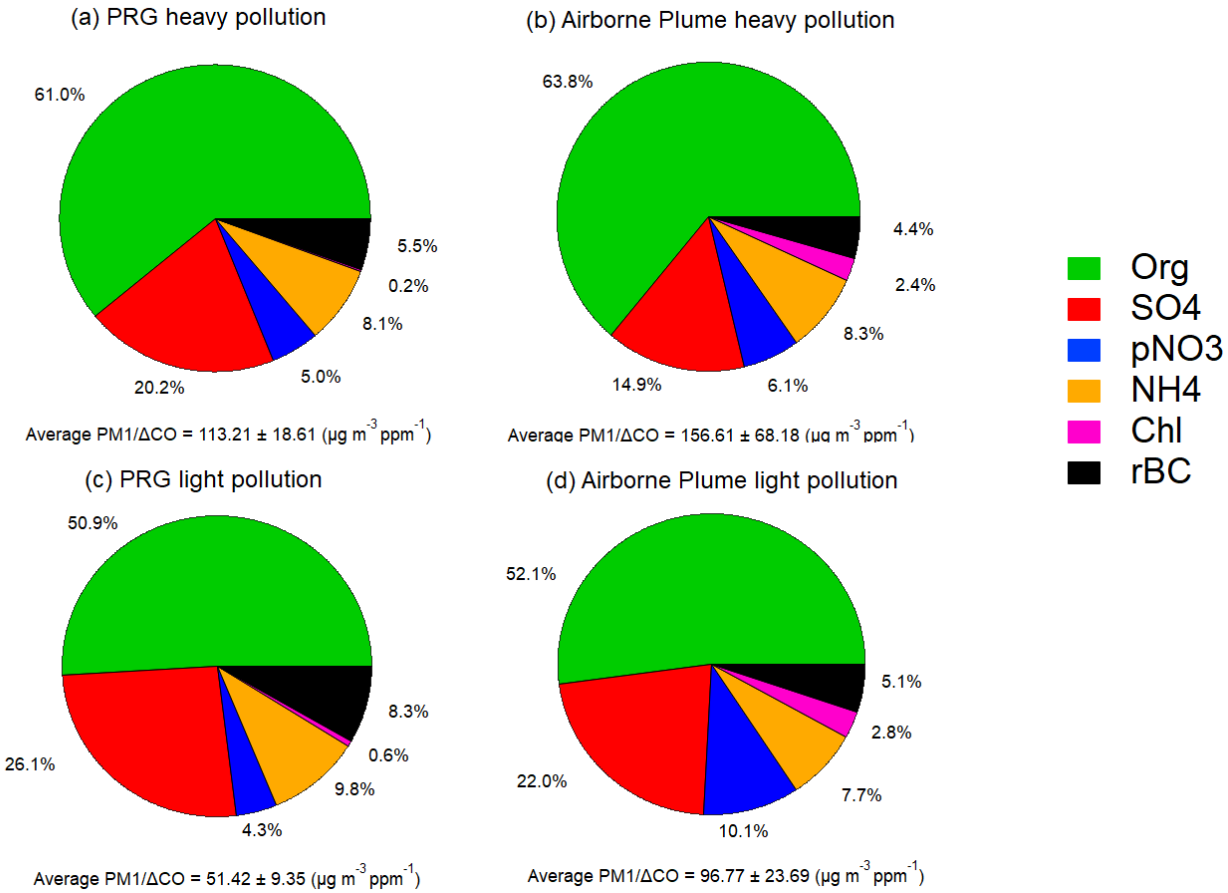

Figure S2: Mass fraction of PM_1 components measured at (a)(c) PRG urban site and (b)(d) downwind urban plume derived from airborne results at heavy and light pollution periods. The uncertainty in the average $PM_1/\Delta CO$ represents the standard deviation.

40 **Unlike Figure 7 in the main text, which includes all flights and the corresponding PRG emission periods, the analysis here is restricted to flights with available SP2 measurements (A033 for the light pollution period, and A028 and A032 for the heavy pollution period), along with their corresponding PRG emission periods.**

S9 Estimation the real part of Org CRI

The real part of the Org CRI (n_{Org}) was retrieved using a volume-weighted linear mixing rule. Mass concentrations of NR-PM_1 species measured by ACSM/mAMS were converted to volume concentrations using densities of 1.27 g cm^{-3} for Org, 1.77 g cm^{-3} for inorganics (Inorg), from which the volume fractions of Org (VF_{org}) and Inorg (VF_{inorg}) in NR-PM_1 were calculated. Assuming linear mixing of CRI, the real part of the Org CRI (n_{org}) was obtained as:

$$n_{\mathrm{Org}} = \frac{n_{total} - n_{\mathrm{Inorg}} VF_{\mathrm{Inorg}}}{VF_{\mathrm{Org}}}$$

where n_{total} is the measured real part of the CRI derived using the method described in Section 2.4 in the manuscript. n_{Inorg} represents the real part of the CRI of Inorg and is assumed to be 1.53, representative of non-absorbing Inorg. BC was not included in this analysis because the airborne SP2 measurements were partly unavailable. Nevertheless, BC contributed only a limited fraction of the total PM_1 mass, with PRG rBC < 10% and airborne rBC < 5%. Therefore, excluding BC is unlikely to affect the observed trend in the variation of n_{Org}. The absolute values of n_{Org} may still be subject to some uncertainty due to other assumptions regarding component densities and aerosol mixing state; here, n_{Org} is primarily used to illustrate the relative variation in the optical properties of Org rather than to provide precise absolute values.

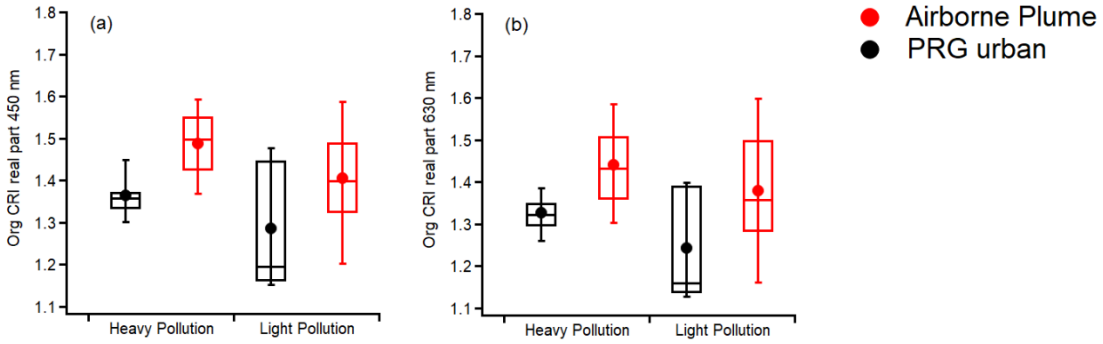

Figure S3: Estimated Org CRI real part values at (a) 450 nm, (b) 630 nm at the PRG urban site and in the downwind urban plume from airborne observations during light and heavy pollution periods.