



Supplement of

Intra-event evolution of elemental and ionic concentrations in wet deposition in an urban environment

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Figure S1 shows the cumulative drop size distribution (DSD) and droplets fall velocity for each rain event. The droplet fall velocity curve, plotted against droplet size, closely resembles the Gunn and Kinzer curve commonly observed for droplets in the range 0.05 to 29 mm (Gunn and Kinzer, 1949). Recorded droplets fall velocity ranges are similar across all events, ranging from 0.15 to 8.8 m s⁻¹. However, there are differences in the number and size of droplets between events. R8, which has a greater rainfall depth and a higher number of drops. On the other hand, R7 is associated with larger droplets ($D_{50} = 0.94$ mm), which explains the higher rainfall rate compared to other events.



Figure S1. Evolution of droplets fall velocity $(m s^{-1})$ and total number of droplets (ND_d) as a function of the droplet diameter (mm) for each rain events, except for R1 for which the disdrometer was not in operation.

REFERENCE

Gunn, R. and Kinzer, G. D.: The terminal velocity of fall for water droplets in stagnant air, Journal of the Atmospheric Sciences, 6, 243–248, https://doi.org/10.1175/1520-0469(1949)006<0243:TTVOFF>2.0.CO;2, 1949.

The precipitation observed by the ceilometer is shown by the yellow-orange to red signals (Figure S2), which shows the precipitation originating from the cloud base and reaching the surface. The signal intensity also illustrates the homogeneity of the atmospheric load on the air column from the surface to the cloud base, except for R5 and R8 where the air columns below the clouds appear inhomogeneous. For example, the measured signal intensity is lower after event R4 (light blue) than before the event (blue-green). This illustrates the effective leaching of the atmospheric column, which is consistent with a decrease of the order of 53% in the PM_{10} concentrations measured at the surface. In addition, for R5, one can also see an aerosol layer between 2 000 and 3 000 m altitude that gradually get down to the cloud level just before it starts raining. Rain events have varying cloud base heights (from 200 m for R6 up to 2 000 m for R8) which, however, can fluctuate within the same event as seen for R8.



Figure S2. Temporal evolution of the signal intensity measured by the ceilometer as a function of time and altitude. The blue and white squares represent the cloud layer base, the black and light blue squares represent the boundary layer height and the color scale corresponds to a proxy of the atmospheric aerosol load. The intense orange-yellow to red lines correspond to the precipitating raindrops.



Figure S3. HYSPLIT 48-h (a-d) and 120-h (e-h) back trajectories of surface and cloud-base air masses for the collected rain events.

Supplement S4



Figure S4. Dust aerosol optical depth (AOD) multi-model simulation provided by the WMO Barcelona Dust Regional Centre from North Africa to Western Europe (<u>https://dust.aemet.es</u>, Basart et al., 2019) for (a) 29 March 2022 at 12h00 and (b) 13 April 2022 at 00h00.

REFERENCE

Basart, S., Nickovic, S., Terradellas, E., Cuevas, E., García-Pando, C. P., García-Castrillo, G., Werner, E., and Benincasa, F.: The WMO SDS-WAS Regional Center for Northern Africa, Middle East and Europe, E3S Web Conf., 99, 04008, https://doi.org/10.1051/e3sconf/20199904008, 2019.

Table S5. Decrease Factor (DF), which correspond to the ratio of the mass concentration of the first fraction to the last fraction, of the particulate and dissolved phases and the ratio of the decrease factor of the particulate to the dissolved phase.

Rain event	DF for particulate phase	DF for dissolved phase	DF particulate /DF dissolved			
R1	28.7	16.5	1.7			
R2	19.9	2.7	7.3			
R3	3.6	1.5	2.3			
R4	49.3	22.1	2.2			
R5	3.6	1.8	2.0			
R6	5.3	1.9	2.8			
R7	10.5	2.6	4.0			
R8	15.0	10.9	1.4			

Supplement S6

Table S6. Washout ratios of elements for the selected rain events R2, R3 and R8

Element	Al	Ba	Ca	Cl	Co	Cr	Fe	K	Mg	Mn	Na	Р	S	Si	Ti	Zn
R2	1 699	827	2 580	3 058	1 278	485	617	1 835	1 449	1 219	1 625	5 291	2 630	2 056	2 492	1 029
R3	4 4 3 6	2 566	9 775	69 618	6 272	3 194	3 806	4 701	7 793	4 733	11 119	16 699	3 223	5 423	7 342	3 792
R8	3 819	1 298	5 472	14 402	917	483	1 555	3 606	4 183	2 009	3 002	2 966	4 444	3 579	3 699	2 327