nature geoscience

SUPPLEMENTARY INFORMATION

https://doi.org/10.1038/s41561-019-0335-5

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Atmospheric transport and deposition of microplastics in a remote mountain catchment

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Supplementary Note 1 - Detailed site description

The study site is located at the Bernadouze meteorological station (OHM Haut Vicdessos Labex-DRIIHM station run by CESBIO and ENSAT INP University of Toulouse[1,2]) (Figure 1). The meteorological station is situated 42°48'14.6"N 1°25'06.8"E, at 1425m a.s.l. within the Vicdessos catchment, in the mountain range of Mid-Pyrenees mountains in south-west of France.

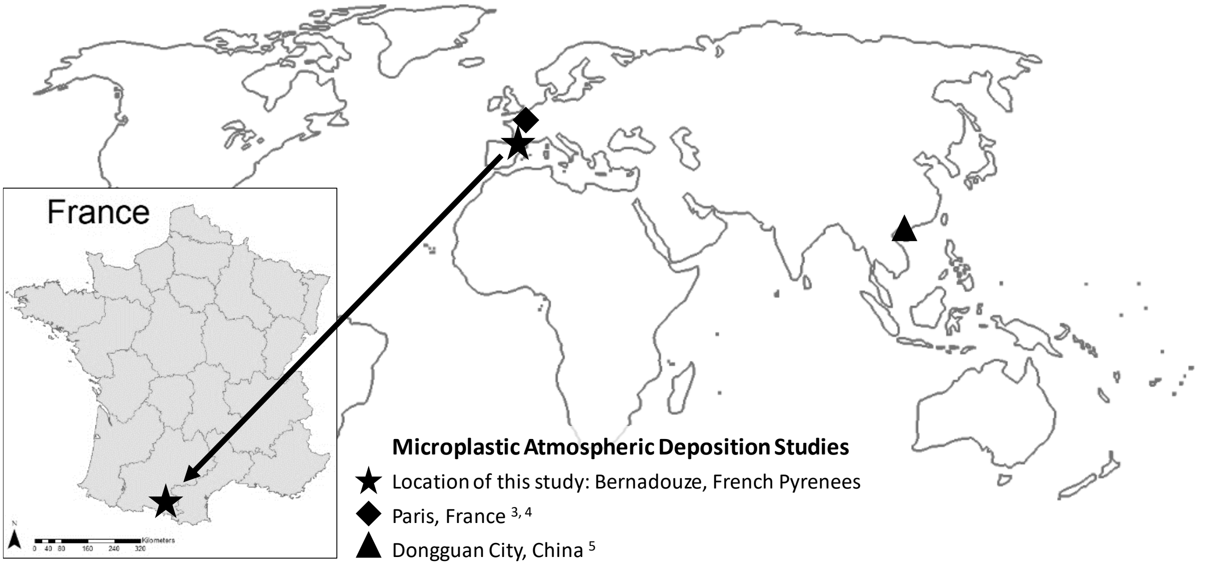

Figure 1. Study site location and previous atmospheric deposition MP studies ([3–5]).

The mountainous regions of the Pyrenees are anecdotally considered a pristine wilderness area due to limited development, difficulty of human access (primarily recreation and agriculture use) and distance from major populations or industrial centres. A review of the literature however shows that the Pyrenees Mountains have been the sentinels of anthropogenic pollution[6–8] as far back as 685AD with lake cores illustrating high levels of lead and remarkable levels of arsenic[9] from mining and industrial activities[10,11]. Pyrenean peat archives illustrate both local and distal pollutant influences; from the Roman Period influence is suggested to occur from the Iberic Peninsula or/and local Bronze Age pollution[12]. The Pyrenees have also been shown to receive bacteria and virus' from airborne deposits of dust from Africa [11,13,14].

The station is located in the saddle of the pass running East-West and is 214m to the south of the only road that traverses through the pass (the road was closed to normal traffic throughout the study period due to standard Winter traffic regulations). The local vicinity is sparsely populated and without industrial, commercial or large agricultural activities and is primarily used for recreational activities (hiking, skiing, environmental education and scientific research). The closest local residential area is a village ~6km to the east in the Vicdessos valley (Vicdessos village, population ~540 [15]) with a moderately sized town located ~25km to the north-east (Foix, population ~9,720 [15]). The field site is approximately 100km south of Toulouse, 120km west of Perpignan (and the Mediterranean coast), approximately 24km north of the metropolitan area of Andorra and approximately 12km north west of the French-Spanish border.

The meteorological station is located in a clearing, free from road access and tree cover. The monitoring location is within a saddle pass that faces east south-east with the Pyrenean mountain range stretching around the site and to the north-west and south-east. The 'Pic de Trois Seigneurs' (2199m a.s.l.) is located 3km to the north north-east, at the top of the adjacent Arbu catchment.

This field site is a location of continuous monitoring for climate and environmental analysis, primarily used in analysis of atmospheric pollution such as Pb, Hg, Sb, CO_2 in conjunction with local peat and mountain lake pollution repositories (studies undertaken by EcoLab, Observatoire Homme-Milieu Pyrénées Haut Vicdessos and Observatoire Midi Pyrenees). For this reason, this location was selected as the preliminary case study site, due to the history of atmospheric pollution research undertaken at this location, the availability of meteorological data and physical sample acquisition.

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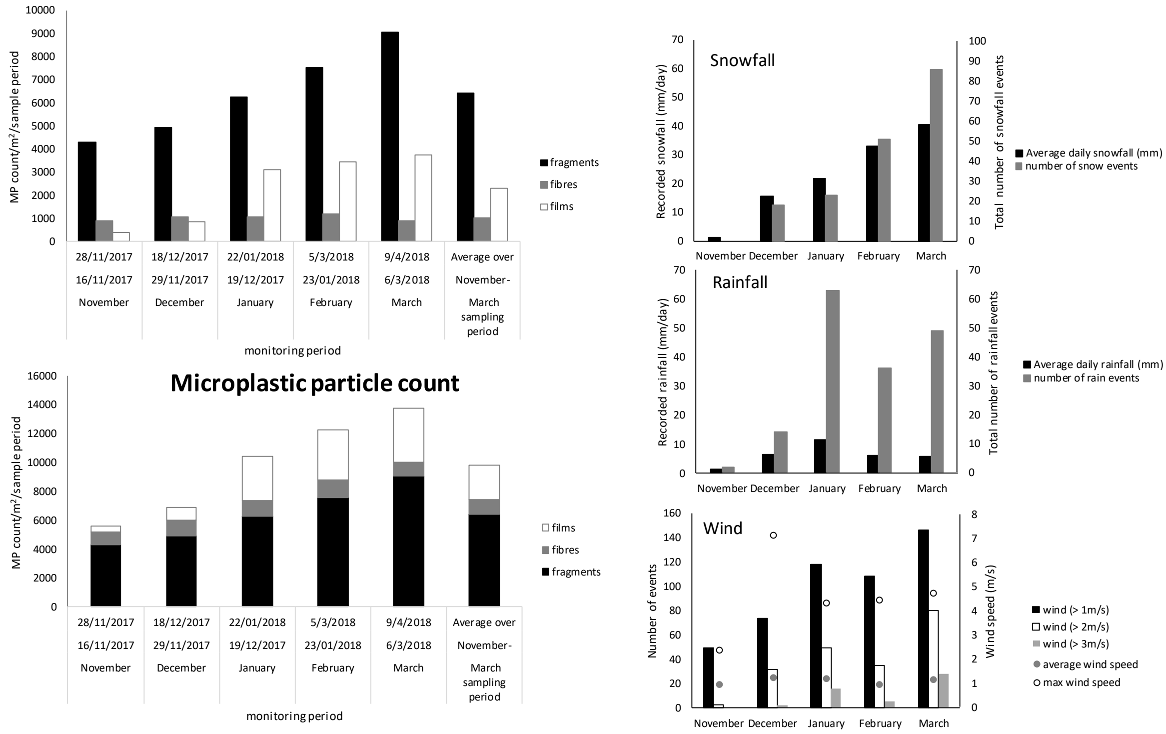
Supplementary Table 1 - Rainfall, snowfall, temperature and wind data relative to microplastic monitoring periods.

					Wind							Rainfall								Snowfall							
	Sampling start date	Sampling end date	Duration of sampling period	Total MP / m2/ day	Average wind speed (m/s)	Predominant wind direction	Maximum recorded wind speed (m/s)	Frequency of wind speeds greater than 1m/s	Frequency of wind speeds greater than 2m/s	Frequency of wind speeds greater than 3m/s	Frequency of wind speeds greater than 5m/s	Total rainfall (mm)	Average daily rainfall (mm/day)	Average rainfall event intensity (mm/hr)	Maximum event intensity (mm/hr)	Number of rainfall events >1mm	Number of rainfall events >2mm	Number of rainfall events >5mm	Number of rainfall events >10mm	Total snowfall (mm)	Average daily snowfall (mm/day)	Average snowfall event intensity (mm/hr)	Maximum event intensity (mm/hr)	Number of rainfall events >1mm	Number of rainfall events >2mm	Number of rainfall events >5mm	Number of rainfall events >10mm
Meteorological data normalised to daily time-step																											
November	16/11/2017	28/11/2017	12	462	0.9	WSW	2.4	4.1	0.2	0.0	0.0		1.5	0.9	11.5	0.2	0.2	0.1	0.1		1.1	0.0	0.0	0.0	0.0	0.0	0.0
December	29/11/2017	18/12/2017	19	359	1.2	W	7.1	3.8	1.6	0.1	0.2		6.5	0.8	8.8	0.7	0.6	0.4	0.3		15.7	0.5	1.3	0.6	0.5	0.4	0.2
January	19/12/2017	22/01/2018	34	306	1.2	WSW/W	4.3	3.5	1.4	0.5	0.0		11.6	0.9	6.4	0.8	0.6	0.3	0.2		21.9	1.0	1.9	0.6	0.6	0.5	0.3
February	23/01/2018	5/3/2018	41	297	0.9	SW	4.5	2.6	0.9	0.1	0.0		6.1	1.5	6.0	0.3	0.2	0.2	0.1		33.1	1.2	2.6	1.2	1.0	0.6	0.3
March	6/3/2018	9/4/2018	34	402	1.1	WSW	4.8	4.3	2.4	0.8	0.0		5.7	1.5	9.2	0.5	0.4	0.2	0.2		40.7	2.0	3.7	2.5	2.5	1.5	0.7
Meteorological data relative to the monitored period																											
November	16/11/2017	28/11/2017	12	5544*	0.9	WSW	2.4	49	2	0	0	18	1.5	0.9	11.5	2	2	1	1	14	1.1	0.0	0.0	0	0	0	0
December	29/11/2017	18/12/2017	19	6819*	1.2	W	7.1	73	31	2	3	124	6.5	0.8	8.8	13	11	8	5	298	15.7	0.5	1.3	12	10	7	3
January	19/12/2017	22/01/2018	34	10401*	1.2	WSW/W	4.3	118	49	16	0	394	11.6	0.9	6.4	27	22	10	6	745	21.9	1.0	1.9	22	22	16	9
February	23/01/2018	5/3/2018	41	12188*	0.9	SW	4.5	108	35	5	0	250	6.1	1.5	6.0	12	10	7	6	1356	33.1	1.2	2.6	51	40	23	12
March	6/3/2018	9/4/2018	34	13680*	1.1	WSW	4.8	146	80	28	0	193	5.7	1.5	9.2	18	13	8	7	1385	40.7	2.0	3.7	85	85	50	23
Total dataset	16/11/2017	9/4/2018	140	58358*	1.1	WSW	7.1	494	197	51	3	980				72	58	34	25	3799				170	157	96	47

* For the dataset presented relative to the monitored period, the MP count is the total MP/m^2 for the total monitored period (i.e. cumulative MP count over the total 34 days for the January sample). For the purposes of this assessment a wind event is defined as an instantaneous wind speed greater than a given threshold. All wind, rainfall and snowfall values are presented *per day* for the 'Meteorological data normalised to daily time-step' and *per monitored period* (i.e. 19 days in November) for the 'Meteorological data relative to monitored period'.

Supplementary Note 2 – Total MP deposition relative to monitoring periods (MP counts without normalisation)



MP deposition for the total monitored periods of November (12 days, 16/11/2017-28/11/2017), December (19 days, 29/11/2017-18/12/20170, January (34 days, 19/12/2017-22/01/2018), February (41 days, 22/01/2018-05/03/2018) and March (34 days, 006/03/2018-09/04/2018).

The non-normalised dataset has been considered and the MP/m^2/monitoring period correlated to actual meteorological conditions specific and recorded for the known monitored durations. Both rainfall and snowfall show moderate to strong significant correlations to MP count in the original (non-normalised) dataset ($r \geq 0.8$, $p < 0.05$). This dataset also presents a strong, positive correlation between the monitoring duration (days) and MP deposition (MP/m^2/monitoring period). The original dataset illustrates a stronger correlation between snowfall (total and number of events) and MP deposition compared to rainfall, suggesting effective snowfall scavenging of atmospheric MP. There is also a trend of increasing daily snow deposition (mm/day) during the total monitoring period (November – March) that corresponds to the (non-normalised) increase in MP deposition trend, while daily rainfall rises to a maximum in January then declines. The frequency of wind speeds >2 m/s and >3 m/s also show positive strong correlations to MP deposition, with higher MP counts for periods recording a greater number of elevated wind speeds.

Supplementary Note 3-Fragment - fibre - film ratio in the mountain atmospheric deposition samples

The composition of MP particles (fragment, fibre, film) is interesting compared to other studies published, generally illustrating lower fibre counts than in previously published city atmospheric deposition studies. There are several possible reasons for the smaller proportion of fibres compared to particles in the deposition samples. These include:

(1) Early MP studies and field sampling campaigns presented plastic fibre counts in environmental samples. There are several studies that present only fibre counts, without corresponding particle or film counts. In early studies this may be partly due to the ease of fibre identification. There may be a resulting expectation of high plastic fibre findings due to the earlier published slight bias towards plastic fibre analysis in environmental samples [1–5]. A Scopus search to identify the number of published articles ("microplastic" AND "environment" AND fibre/film/particle/fragment) identified a total of 525 articles on microplastic and environment, 169 discussed fibres or microfibres specifically (322 - particles, 101 - fragments, 31 – films). It is also noted that Shim et al.[6] presents several studies of sea water and sediment that show fibre counts similar and smaller than that found in this study (with acknowledgement that these are different environmental compartments).

(2) It is potentially possible that in microplastic analysis to date there is a limited identification and description of the very small particles in previous studies due to particle size analysis limitations (for example, studies may consider plastic greater than 50 or 100 µm). It has been suggested through several studies[7,8] that the particle count increases exponentially with decreasing size. Therefore, potentially, the ratio of fragments to fibres may be different in this study due to the analysis of MP particles down to a lower size limit than some previous studies. When only >200µm MP particles are considered in this remote mountain field analysis, the proportion of fibres in the sample rises to 70%, comparable to the higher fibre counts presented in the mega-city studies (Paris and Dongguan)[3,9].

(3) The deposition collection method is open to the atmosphere for the duration of the sample period. While films and fragments may become and remain deposited in the standard field rainfall samples the fibres are potentially lighter and more easily entrained and therefore if dry deposition occurs the potential for resuspension may be greater for these MPs than for films or fragments. However, the entrainment potential of fibres vs films vs fragments has not been tested (in the field or laboratory) and so this potential field fibre re-entrainment cannot be evidenced.

(4) The sample preparation methods follow standard protocols previously used and published. It is noted however that the use of H_2O_2, even when diluted, can results in 'loss' of fibres through acid dissolution. This could potentially affect aged or degraded fibres or fibres of Rayon or similar composition[10,11], resulting in a lower fibre count in comparison to fragment numbers. It is noted that the Fenton's reagent method[10] may assist in limiting sample fibre loss during organic material removal.

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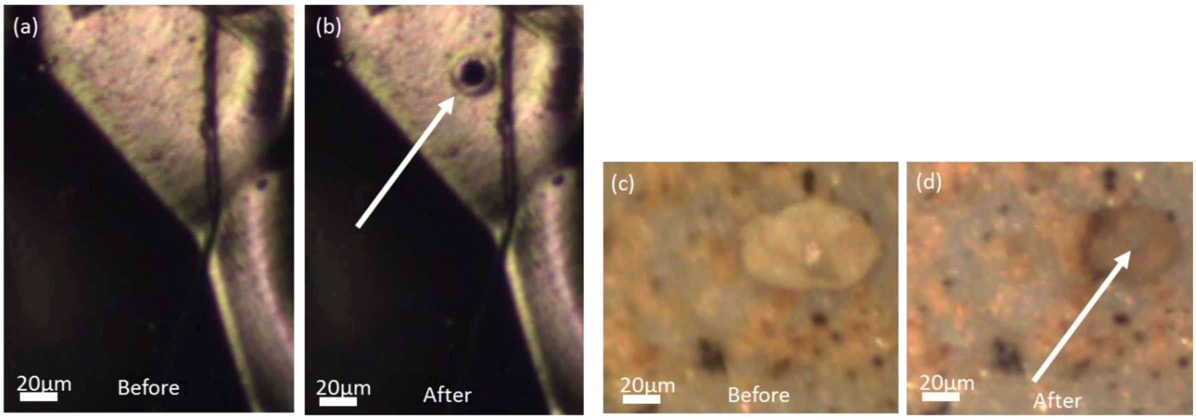
Supplementary Note 4 - Images of Raman laser impact on plastic



Images of Raman laser impact on plastic at 50% (a, b) and 100% (c, d) laser power (a and b is a control plastic particle, c and d is a sample fragment). Plastic becomes vaporised (burns) leaving a hole at the focal point of the laser when high laser power is used for analysis.