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**SIMULATING THE DISPERSION OF MICROPLASTICS IN THE ATMOSPHERE
TOWARDS A REMOTE SITE**

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Abstract: The study of the dispersion of microplastics in the atmosphere represents a pioneering field of research, a very few works on this subject can be found in literature. In this work, the potential long-range dispersion of microplastics in the atmosphere is investigated using the Lagrangian stochastic particle model MILORD. The rationale is to apply MILORD model in the backward mode, in order to identify possible emission sources that may determine the microplastics pollution in sites or areas of interest. The sensitivity to the resolution of the meteorological input, to the diffusion parameters and the effect of the settling velocity values have been primarily investigated. A case study has been chosen for reference, concerning the observations of atmospheric microplastics deposition in a remote and pristine site in French Pyrenees, for which the long-range dispersion may play a main role. Preliminary results of simulations for different periods and different model configurations are presented and discussed.

Key words: *atmospheric microplastics; backward dispersion; long-range; MILORD model; numerical simulation.*

INTRODUCTION

In 2015 for the first time the microplastics total atmospheric fallout, mostly composed by fibres, was highlighted in Greater Paris (Dris et al., 2015), some years later González-Pleiter et al. (2021) presented the first direct evidence of the microplastics presence above the planetary boundary layer. In the meantime, several studies sampled and reported the atmospheric microplastics deposition in different places: many cities, such as London (Wright et al., 2020), Hamburg (Klein et al., 2019), Dongguan (Cai et al., 2017), some remote and pristine area, as the French Pyrenees (Allen et al., 2019) and the Tibetan Plateau (Zhang et al., 2021), and in the ocean too (Liu et al., 2019; Trainic et al., 2020). The unequivocal presence of microplastics, hereafter MPs, in the atmosphere highlighted the importance to thoroughly investigate all the possible transport pathways in order to properly address the problem, using advanced tools like numerical models. Modelling MPs dispersion in the atmosphere is still a pioneering research field, so there is not an established reference framework yet. Hence, with the aim to contribute building a useful framework, in this work the long-range atmospheric dispersion model MILORD is applied to investigate the effects on microplastic transport of some key parameters, such as the settling velocity, at the same time assessing the model sensitivity to the resolution of the meteorological input and to the value of the diffusion coefficients. The case here under study refers to the work of Allen et al. (2019), where the deposition of MPs in the remote site of Bernadouze in the French Pyrenees and their possible atmospheric pathways were investigated.

SENSITIVITY ON RESOLUTION AND DIFFUSION PARAMETERS

MILORD simulates the transport, diffusion, removal and deposition of tracers on the long-range, that is at the planetary scale. MILORD is grid-free, being a Lagrangian model, and the dependency on the grid resolution regards only the meteorological datasets used in input. In previous studies a resolution of $0.5^\circ \times 0.5^\circ$ of the ECMWF analyses was used for the input files to address long-range dispersion (Boetti, 2015; Greco, 2018), a grid spacing that is still characteristic of large scale simulations. Clearly, this horizontal resolution can be too coarse to properly resolve the orography, in particular in complex terrain. For example, in the considered case study (Allen et al., 2019), the altitude of the Bernadouze meteorological station is 1425 m whereas the height obtained by the ECMWF files, interpolating the values over the $0.5^\circ \times 0.5^\circ$ resolution, is only about 640 m in the surrounding of the site.

Focusing on the orography, its altitude is determined for each particle location and at every timestep, hence an interpolation method is necessary to extrapolate the orography out of the values at the grid-points of the ECMWF input files. In MILORD, a bilinear function is used to assign the height of the orography at the particle location, interpolating the values available at the four grid-points of the grid-cell to which the particle coordinates belong. Clearly a coarse horizontal spatial resolution leads to a smoothing of the orography that can affect the representativeness of the real altitudes and it may influence the particle trajectories computation. The meteorological variables are also input at the same resolution; therefore, smaller scale circulations may be difficult to be captured. For these reasons, to assess the sensitivity of the model to the resolution of the input fields, ECMWF meteorological fields at the finer horizontal scale of $0.25^\circ \times 0.25^\circ$ were used for the first time in the model MILORD, in addition to the original $0.5^\circ \times 0.5^\circ$ set; for each resolution 11 pressure levels were considered from the ECMWF analysis files: 1000, 925, 850, 700, 500, 400, 300, 250, 200, 150, 100 hPa.

As regards the diffusivity, in previous works (Desiato, 1998; Boetti, 2015) it was proven that the tracer dispersion in MILORD simulations is well-captured using large values of the diffusion coefficients, $K_x = K_y = 15000$ and $K_z = 150$ m^2/s (Set I). Here, the efficacy of those values was tested also for the case where a finer resolution of the input meteo fields is used. Furthermore, for both resolutions of the ECMWF input, the model sensitivity was evaluated using a second set of value for the diffusion coefficients, $K_x = K_y = 1500$ and $K_z = 50$ m^2/s (Set II), largely varying their magnitude, so to appreciate potential differences in the spread of the plume of particles. The simulations were run in backward mode for a 13-days period in November 2017, the retro-emission starting on 28/11 at 00:00 and ending on 16/11 at 00:00. Assuming that MPs particles in the French Pyrenees site arrived continuously through the atmosphere, the retro-emission was continuous and for each timestep one particle was retro-emitted so to easily track single trajectories.

A first interesting achievement was the improvement in the reliability of the orography values using the $0.25^\circ \times 0.25^\circ$ resolution: the corresponding height interpolated at Bernadouze site was about 1450 m instead of 640 m. Since here particles arriving at the receptor are traced, the height of the emission is assigned at the surface, referring to the height of the orography and corresponding to pressure values of 960 hPa and 855 hPa, respectively. The 3D plots in Figures 1 and 2 represent the plume of particles moving in the domain in the 13-days simulation period, for the two resolutions $0.5^\circ \times 0.5^\circ$ and $0.25^\circ \times 0.25^\circ$, respectively. By the comparison between the 3D-plot, outlining the particles height distribution, it is possible to observe some difference in using Set I instead of Set II for the diffusion coefficient values. As expected the lowest diffusion coefficients leads to a lesser vertical diffusion. The lower diffusivity has an effect also on the horizontal, allowing particles from larger distances to travel further and reach the receptor. The 3D-plot reported in Figure 2, which used the $0.25^\circ \times 0.25^\circ$ resolution, show a similar behaviour and analogous differences when varying the diffusion coefficients. Even using diffusion coefficient values orders of magnitude lower than the usually adopted do not affect significantly the particle dispersion since the main distribution path remains. The evidence reported by Desiato (1998) and Boetti (2015), assessing that the high values of diffusivities are more representative at these large scales, remains confirmed also when using the finer resolution of $0.25^\circ \times 0.25^\circ$; thus, the Set I is maintained to model the diffusion of MPs in the long-range dispersion.

A large variation appears comparing the 3D-plot obtained with the different resolutions, comparing Figure 1 and Figure 2: with the finer one, $0.25^\circ \times 0.25^\circ$, more MPs are found to reach higher altitudes and the shape of the area covered by the particles in the 13-days simulation shows some difference, yet overall, compared to the coarse one, the particles well depict the core of the dispersion region; these differences can be ascribed to the differences in the ECMWF wind fields at the two resolutions.

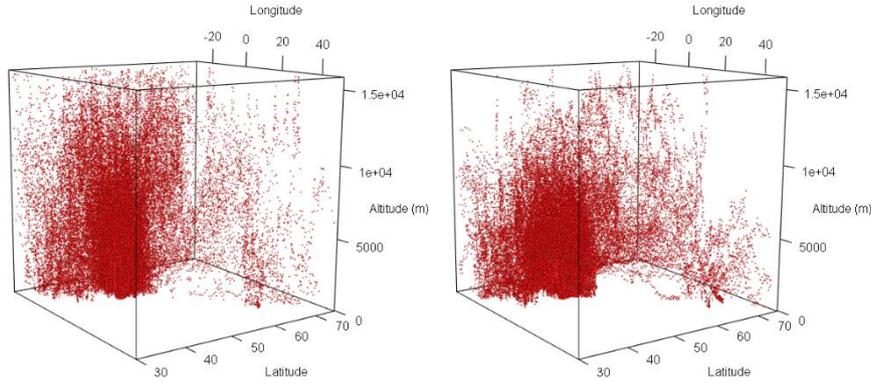


Figure 1. The 3D-plots illustrate the particles height distribution referring to the November period using horizontal resolution of $0.5^\circ \times 0.5^\circ$; considering SET I (left) and Set II (right) diffusion coefficients.

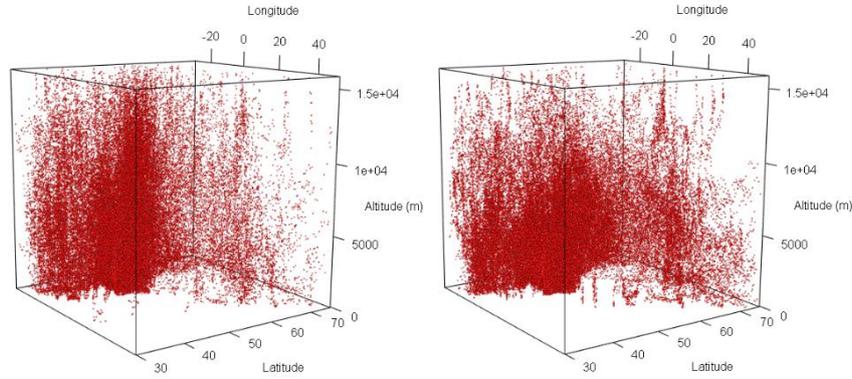


Figure 2. As in Figure 1 but for the horizontal resolution $0.25^\circ \times 0.25^\circ$ for the meteorological input field

INFLUENCE OF THE SETTLING VELOCITY ON DRY DEPOSITION

A first study on the influence of different settling velocities on the dry deposition was conducted. Four different runs were performed using four different deposition velocity values, based on the literature (Table 1), for three different periods of the year (November 2017, January and March 2018), and for each resolution, to assess the possible influence of the velocity on MPs dry deposition.

Table 1. Settling velocity values reported in the literature until 2020

Reference	Settling Velocity (ms^{-1})
Allen et al. (2019)	0.1
Wright et al. (2020)	0.06 (fibrous MPs)
Trainic et al. (2020)	0.32 (non-fibrous MPs)
Allen et al. (2019)	0.001

The computation of the dry deposition in MILORD is done by a subroutine that calculates it at each time step if the particles is inside the Planetary Boundary Layer, hereafter PBL, based on the following formula:

$$D_{dry} = N_0(1 - e^{-\lambda_d \Delta t}) \quad (1)$$

where N_0 is the initial pollutant amount and λ_d is the dry deposition coefficient defines as:

$$\lambda_d = \frac{v_d}{H(x, y, t)} \quad (2)$$

with v_d representing the deposition velocity and $H(x, y, t)$ the particle height.

The calculation in the backward mode has been modified so that the reverse modelling reconstructs the amount of pollutant that the particle would have lost due to the depletion processes, before reaching the sampled site (receptor). For this investigation, the simulations considered a period of one day in which the back-trajectory of one particle was followed and only the settling velocity value was modified from one

simulation to another. The analysis here presented refers, as example, to the November case study: the single particle is retro-emitted on 26/11/2017 at 00:00 and the simulation of the backward trajectory lasts one day, starting at 00:00 on 26/11/2017 and ending at 00:00 on 25/11/2017.

For each resolution, two graphs were elaborated and compared:

1. The trajectory map; here the colour scale represents the amount of MPs carried by the particle in base-10 logarithmic scale. (Figures 3).
2. The amount of MPs transported by the particle as a function of the distance it covers; here the line-graphs outline the influence of the settling velocity on the dry deposition: the vertical axis shows the amount of MPs carried by the particle, whereas the horizontal axis represents the distance travelled by the particle (Figures 4).

By the comparison of the trajectory maps (Figure 3) a variation emerges in the path followed by the particle considering the two resolutions. This is reasonable since, with the finer resolution, the interpolated wind field manage to better capture smaller circulation structures. Furthermore, for the case of November, a remarkable difference in the amount of MPs transported appears comparing the simulations with different resolutions: with the finer one the particle spends less time inside the PBL, hence it “acquires” a lesser amount of MPs due to the dry deposition. Moreover, Figure 4 shows that when considering the highest settling velocity of 0.32 ms^{-1} , a particle should carry a much greater amount of MPs arriving from distance greater than 250 km, making this velocity a less probable value for the long-range transport compared to the others.

In this single-particle case, the difference in the MPs amount is due to the fact that when travelling close to the PBL height, even small differences in the value of the wind field and turbulence, determining the displacement of the particle, and in the PBL height daily development, may let the particle move out/in the PBL layer. Only when inside the PBL, the particle gains back the ‘dry-deposited’ amount in the backward mode, thus leading to potential differences in the particle MPs mass. Concerning the trajectories, it is important to notice that in these simulations only one particle was considered and back-tracked, while in the Lagrangian modelling approach a great number of particles must be released and traced in order to statistically describe the actual path of the plume. In this sense on average the paths followed by the particles are in good agreement for the simulations corresponding to the two resolutions of the input fields.

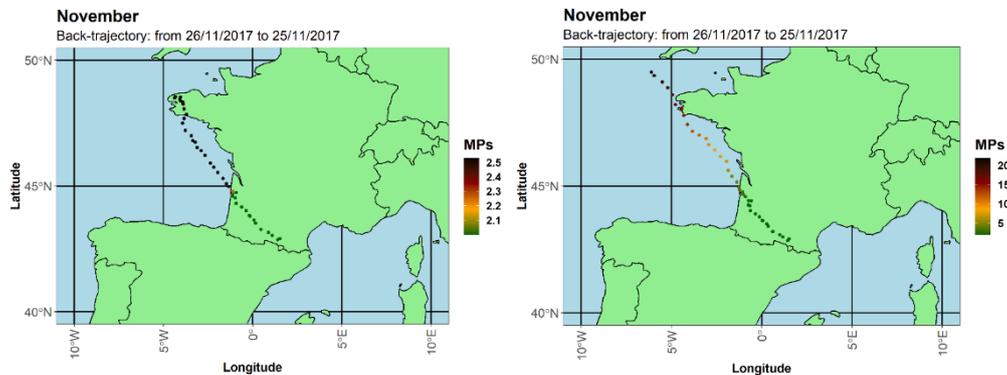


Figure 3. November case. One particle retro-emitted the 26/11/2017 and the followed daily back-trajectory, using the meteorological input fields with $0.5^\circ \times 0.5^\circ$ (left) and $0.25^\circ \times 0.25^\circ$ (right) grid resolution.

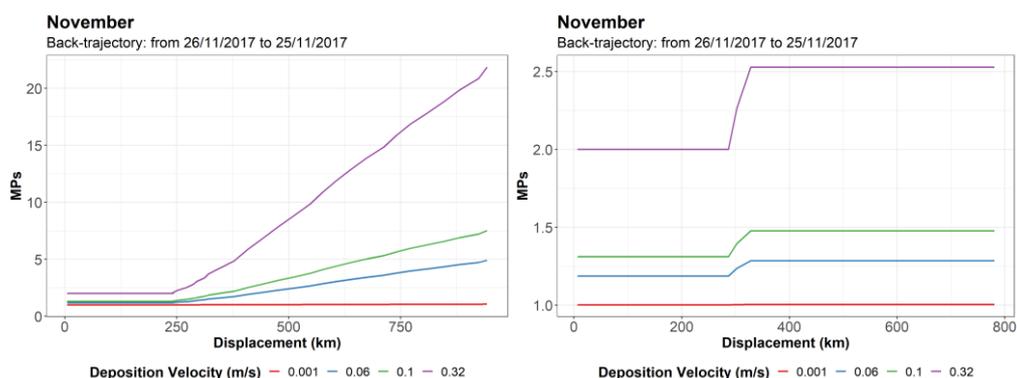


Figure 4. November case. Amount of MPs carried by the particle as function of the travelled distance from the receptor site, using the meteorological input fields with $0.5^\circ \times 0.5^\circ$ (left) and $0.25^\circ \times 0.25^\circ$ (right) grid resolution.

CONCLUSIONS

Even in the difficulty to obtain quantitative results, an important first study was conducted comparing four different settling velocities, based on the reviewed literature, to assess their influence on the dry deposition in the long-range transport. The less likely value for the settling velocity characterizing the long-range transport was found to be 0.32 ms^{-1} , reported by Wright et al. (2020) to describe non-fibrous microplastic.

Moreover, the influence of settling velocities was also evaluated using the two different resolutions of the meteorological fields, $0.5^\circ \times 0.5^\circ$ and $0.25^\circ \times 0.25^\circ$. The investigation delineates a remarkable changing, comparing the two resolutions, in the amount of MPs transported by the particle in order to arrive at the receptor; further investigations will be necessary regarding the finer resolution in order to understand if some changes in the parameterization of the physical processes will be required.

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