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### **Inferring microplastics origins in the Mediterranean Sea by coupling modelling and** *in-situ* **measurements**

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### **Abstract:**

This work focuses on the dynamics and retention of microplastics (MP) in the Mediterranean. MP manta-net trawls were performed in autumn 2019 north of the Balearic Islands and along the Balearic Front (BF). Lagrangian modelling was used to find the MP collected origin during the campaign. These combined results show that North of Mallorca is a temporary retention zone of 3 months variability, with MP origin being the Northern Current (NC) and the Gulf of Lion (GOL). Retention processes were less clear along the BF, due to frontal dynamics together with the strong northerly winds. However, it appears that the origin can differ between the North (i.e. the large North-Westerly basin, including the GOL and the NC path) and the South (short distances around the zone) of this front. In both areas, the wind and the current variability are strongly conditioning the existence and position of the MP concentration zones.

### **1. Introduction**

Global production of plastic materials increased twenty-fold in the last sixty years and has reached the impressive figure of 367 million tons in 2020

(https://plasticseurope.org/knowledge-hub/plastics-the-facts2021/). Despite the past Covid period for which production and demands did slow down, the plastic production is still expected to grow for the next decades. As a result of such ubiquity in everyday uses, 275 million tons of plastic litter were generated in 2010 by coastal countries [Jambeck et al., 2015]. Among this production, it is estimated that between 4.8 to 12.7 million tons have been transported in different ways into the nearby oceans or seas [Jambeck et al., 2015]. Eriksen et al. (2014) estimated that 92 % of all plastic items found at sea were smaller than 5 mm. These small particles, referred to as microplastics (MP), can either result from the breakdown of manufactured objects or can directly enter the marine environment as granules,pellets or fibers. Several recent review papers underlined the impact of MP

at several levels of the trophic chain (Suman et al. 2021, MacLeod et al. 2021, Elizalde-Velázquez et al. 2021), highlighting the numerous threats brought by plastic in the ocean.

The Mediterranean Sea has been shown to host similar or even higher MP concentration to the major large scale accumulation zones of plastic, such as the giant garbage patch in the North Pacific [Cozar et al., 2014]. However, when it comes to find offshore retention zones, few in-situ works have been able to demonstrate their existence [Van Sebille et al., 2015]. Nonetheless, the work from [Mansui et al., 2015], although being purely numerical, was one of the first to demonstrate statistically that large-scale zones could be evidenced as retention magnets in the Mediterranean basin, but that these areas were very variable in time mostly. Indeed, the study shows that some zones are temporary retention areas but cannot be strictly defined as permanent. The main cause is the high seasonal variability of the Mediterranean Sea dynamics, featuring significant mesoscales activity under the form of eddies and frontal dynamics. One limitation of this work was that although being at a relative high resolution compared to former works, the resolution was still too coarse to allow spotting very fine scales retention areas due to frontal dynamics for example.

The main objectives of this work are : (i) evaluate the differences in retention dynamics between two relatively close areas in the Mediterranean Sea, both featuring mesoscale dynamics. The chosen sampled areas were North of the Balearic Islands and a frontal East-West zone between Menorca and Sardinia islands; (ii) improve knowledge on the origin of retention zones in the Mediterranean Sea to provide better information on the MP fate as well as possibly help the set-up of numerical simulations especially regarding the initial conditions.

In order to complete these purposes, a one-month oceanographic campaign during fall 2019 has been performed on both selected zones to collect surface MP with a regular Manta net trawl protocol. The first location has been selected as former campaigns have shown high MP concentration is this area [Cozar et al., 2014]. The second area was chosen because it is a frontal zone for which the Atlantic Waters that spread into the Algerian Basin are meeting the saltier and colder waters of the Liguro-Provençal area, leading to frontal dynamics between water types of therefore different histories and origins. For a full description of this frontal system please refer to [Barral et al., 2021]. In addition, the statistical numerical modelling work from [Mansui et al., 2015] has also been used to determine the likely origin of the areas featuring the high MP concentrations observed in both zones during this campaign. To do so, an ensemble of computations has been done by selecting the particles at the areas of the sea campaign, and then by tracking them back in time (for specific time duration) along their trajectories (already computed) , to identify their geographical origins.

# **2. Material and method**

# 2.1. Sampling sites and MP collection

The Oceanographic cruise has been conducted from mid-September to mid-October 2019 with the research vessel « Septième Continent » belonging to the NGO « Expédition Septième Continent ». MP were collected at sea using a manta trawl (mesh size  $300 \mu m$ ), using a mechanical flow meter (Hydo-bios, Kiel, Germany) fixed at the center of the aperture of dimensions 0.25m x 0.50 m, in order to measure the volume sampled. Wind speed and sea state were recorded in order to apply a correction on the amount of MP collected, following the algorithm adapted by [Poulain-Zarcos, 2020].

MP were recovered at 13 stations in two separate areas as shown in figure 1: one in the North of the Balearic Islands and one East of Menorca. After careful rinsing of the net with water from the sampling site, macro-debris of natural origin (algae, leaves,...) were eliminated after thorough rinsing above the collector. In order to analyze all the MP collected, the sample was transferred on 300 µm mesh size metallic filters then stored into ziplock bags and frozen at -20°C. In the laboratory, MP sorting and numbering were carried out classically and described in [Poulain et al. 2019]. Briefly, a first sorting of MP has been done without digestion to extract the larger pieces. In order to extract most of the plastic, the remaining samples containing eventually remaining MP and organic matter were subjected to chemical digestion (10% potassium hydroxide for 48 hours at 40°C). The remains of the digestion were dispersed in glass petri dishes and the MP were then sorted out under a binocular microscope by placing the petri dish successively on a white, black and red piece of paper in order to isolate all the MP whatever their colors. The extracted MP with and without digestion were manually transferred to a petri dish, a photo was taken and the size of the MP was determined by contour detection using Image J®. We used a correction factor to evaluate the MP concentration (number of particles per cubic meter) by using a wind-driven vertical mixing correction model taking into account individual particle properties (dimension, shape and density) as described in [Poulain et al. 2019].



*Figure 1: Location of the 13 stations (black dots) for which manta net trawls have been performed. Each number close to a black dot represents the station number (cited in table 1). The background is the averaged SST ( ◦C) over the campaign period (25/09 to 09/10/2019) derived from the copernicus marine service reanalysis MFS [Escudier et al., 2020]. Stations 9 and 13 are superimposed.*

### 2.2. Numerical methods

2.2.1. Statistical Lagrangian forward simulations from Mansui et al. (2015).

The drift simulation of the particles at the oceanic surface was performed in two stages: first, the ocean state and the velocity fields were computed with an appropriate ocean general circulation

model (OGCM). Second, the drift of the virtual particles mimicking the MP was computed by an advection model using the velocity fields computed from the OGCM. To do so, the NEMO OGCM was used, set as a regional configuration named MED12 [Lebeaupin-Brossier *et al.*, 2012].The domain was defined on a 1/12 degree ORCA grid [Gurvan *et al.*, 2017] with 567 by 264 grid points covering the entire Mediterranean Sea. This MED12 configuration was forced by the ALADIN-Climate atmospheric fields [Herrmann, 2011] from the METEO-France center, with 3 h intervals and a 10 km by 10 km grid. The resulting ocean model configuration allowed to solve the mesoscale circulation, with a fully resolved wind driven circulation, and was used in numerous studies ([Beuvier et al., 2012], [Waldman et al., 2017], [Waldman et al., 2018]). Then an advection model named ARIANE has been used to transport the MP using the velocity fields generated with MED12. This tool is an open-source code [Blanke and Raynaud, 1997] available at http://www.univbrest.fr/lpo/ariane.

In order to determine the likely area of origin of the different sampled stations, a large ensemble of ARIANE simulations have been processed, using the work that have been preliminary conducted by [Mansui et al., 2015]. The basics of this ensemble experiment was thoroughly explained in this widely cited reference but one can recall the main points of the created database. The velocity fields available were daily averaged files from January 2001 to December 2010. Every day, particles initially equally spaced every 10 km (in the zonal and meridional directions) were released, and advected for a duration of 1 year, for each available day of the database. The total number of 1-year simulations was 3287, all with the same spatial homogeneous initial deployment, but with a temporal initial condition delayed by 1 day. The position of each particle along its trajectory was saved every day. In other words, one-year ARIANE simulations was performed every day from the 1st of January 2001 to the 31st of December 2009, and for each of these 3287 runs, the complete trajectory (i.e. daily position) of each of the 25,500 particles has been saved. The obtained results have been used for several statistics such as identifying the main open ocean retention areas or the main beaching areas [Mansui et al., 2015].

# 2.2.2. Backward in time identification and counting

For the present study, the originality lied in using this already created database as a source of information for identifying the initial distribution areas of MP that were present during the Autumn season in the zones of the sea campaign. Therefore, the present methodology was as follows: first, the particles located in the areas corresponding to the sea campaign were selected. Second, only the particles present in these areas during the autumn season were kept, as it was the season of the sea campaign. Third, by going back in time along each selected particle trajectory, all the initial positions were localized and counted. In Mansui et al. (2015), the typical advection time reaching

the main patterns of accumulation was obtained after 3 months and it was shown that one year transport was enough to strengthen the 3 months patterns obtained. Therefore, the tracking along the trajectories has been made in priority for 3 months back in time but also for 6 and 12 months. Finally, the results presented are normalized by dividing the counted presence for one location with the location presenting the maximum of occurrence, giving a range between 0 and 1. Other normalization were not efficient due to the very large difference in particle loss during their trajectories, due to stranding, and depending on the region, the seasonality and the inter-annual variability. Extra-information regarding the lay-out of these results are given in section 3.4.

# **3. Results and discussion**

#### 3.1 MP concentration from the oceanographic cruise

As detailed in section 2.1, the MP concentration were obtained after visual sorting of each piece collected with the manta trawl and calculated using sea-state corrections from Poulain et al. (2019). With this method, inferior and superior estimations of the corrected concentrations are obtained, mainly derived by first considering uncertainties on the MP shape and buoyancy, and second by solving the flux balance between the turbulent flux and the buoyancy one (Kukulka et al, 2012). Table 1 shows that MP were collected in quantity at all stations, but with a large range of variation between 4 to several hundred thousand of pieces per  $km^2$ . The displayed SST is derived from CTD casts performed at each station. We have chosen to keep this indication as SST can also be used by the reader to roughly locate the stations: below 23.5 to 24 °C, the station is located in" cold" waters so North of the frontal zones, and above this value it is likely to be located in the" warm" waters so South of the frontal zones.



*Table 1: MP concentrations at sampled stations during the sea campaign. (NB) stands for North of Balearic Islands and (BF) for the Balearic Front. MP inferior and superior estimations in thousands of pieces per km<sup>2</sup> after [Poulain-Zarcos, 2019] sea-state correction. The SST is extracted from CTD casts performed at each station. The stations in the NB zone are printed in red for extrareadability.*

### 3.2. Focus on the North Balearic (NB) zone

The highest MP concentrations was recorded in the North of the Balearic Islands, but at a varying distance between the Spanish continental coast and Mallorca Island depending on the date (Stations 2, 3, 4 and then 15, 16, 17, Figure 1 and Table 1). This supported the idea that the current structure in this zone is favoring at least a temporal accumulation in the area (Figure 2), representing the numerically modelled averaged surface velocity over year 2019, from the CMEMS reanalysis. A branch of the Northern Current (NC) which is the major current of the North-Western Mediterranean zone is turning east before reaching Majorca and then partly feeding the Balearic current that flows south-east. In the center of this large meander, a small eddy-like pattern is highlighted by the yearly averaged from the CMEMS analysis. This eddy-like pattern can be acting as a retention structure for plastic, but as discussed by [Mansui et al., 2015], it can probably not be considered as a stable structure, but more a weather and seasonal dependent structure. For instance, [Ananda et al., 2002] have showed that this structure can hardly be considered as a regular eddy: they explain that a large and long lasting, clearly identified anticyclonic eddy has been spotted from September 1998 to March 1999, but such a long-lasting event is not permanent and rather rare. However, this also proves that such eddy can last during the fall season, and in this case favoring plastic retention as discussed more recently in [Aguiar et al., 2022] for which at least two eddies of 2 to 4 months life have been identified in the area in 2010 and 2017. As for more regular conditions, it can be inferred that the frontal dynamics at the offshore boundaries of both currents (Northern current and Balearic current) are anyway helping the accumulation of floating material in this area [Suaria et al., 2021].



*Figure 2: Surface velocity averaged over year 2019 (in m/s). Numerical data from CMEMS reanalysis [Escudier et al., 2020].*

As the area is also a frontal zone of strong currents [Ananda et al., 2002], the spatio-temporal variability of the current and frontal structures must be strong. This might explain why, stations 4 and 16 very close geographically, but sampled 13 days apart, gave very different results (low for

station 4 and high for station 16). Similarly, stations 2 and 15 (conducted 14 days apart) are very close but with a high concentration for the first and low for the second. One explanation could be that the current and weather conditions have moved north the high retention area in this two-week period, shifting this hot spot initially about 40.3° N to 40.7°N, so about 50 km northward. Indeed, if the wind conditions are investigated, it turns out that for the NB area, the wind has been blowing mostly from the South, with an average intensity of 5.1 m/s (see supplementary material for more information). This tells us that the accumulation zone could have been pushed northward between our first and our second visit in the zone. Another possibility is that the high retention area is intermittently evacuated by the currents and that a new high retention area is created, but with a significant variability in the accurate location of the hot spot.

### 3.3. Focus on the Balearic Frontal (BF) zone

For the BF zone the evidence is less clear as during the period, several storms (North-Westerly wind mainly, c.f. supplementary material) have also moved and mixed the frontal zone. Hence, the replication of the trawls at the same locations did not give similar results, as also noticed for the NB zone, but the concentration rates did not give large gradients either. Again, if the wind conditions are considered, it appears that North and North-Westerly winds have been blowing more or less strongly during the entire campaign, with a classic fetch pattern for the region. One example of such fetch, based on the MeteoFrance operational ARPEGE model, is available in the supplementary material. The wind study shows that the BF area has been strongly subjected to the northerly wind regimes of this sub-basin. More precisely, the daily averaged wind strength over the area calculated from ARPEGE shows that the wind has picked up quite strongly between the 2<sup>nd</sup> and 3<sup>rd</sup> of October and then the 7*th* of October. These strong winds might have completely changed the frontal zones and dynamics, in all sampled areas. Indeed, if we assume that stations 7 to 9 are nearly equivalent to stations 11 to 13, the main difference is that for the first period the wind has been quite low the days before (around 4.5 m/s) and for the second period, the wind reached strong values up to 8.8 m/s. Now, when looking at table 1, it appears that the increasing gradient towards higher values have been moved south, which is intuitively in good agreement with Northerly winds pushing south the frontal zone and therefore pushing south the MP accumulating along these fronts. This second hypothesis is however difficult to prove since no extra *in-situ* data of MP measurements exists over the area with a sufficient replication and the actual data remain of low magnitude compared to the NB zone. Only significant drifters deployment in the area could really help proving this possibility, but to our knowledge, two few examples are available.

Hence, in order to complement the understanding of the collected *in-situ* data distribution, next section focuses on the potential identification of the short to long range origin of the sampled areas by means of numerical methods.

# 3.4. Numerical identification of the origin for the collected MP

Due to the uncertainty of the numerical simulations coming first from the OGCM currents (more precisely its horizontal resolution), and second from the Lagrangian simulation itself, it has been chosen to track particles back in time from rectangle domains including a group of stations instead of focusing on each sampled station individually. Two boxes have then been studied, one including the stations located in the NB zone and the other in the BF zone. For the presented figures in the next sections, please note that the color bar represents the counted particles (originating from the chosen box) plotted for each initial position and divided by the maximum counted presence obtained for each map. This numbers are widely varying between the 3, 6 and 12 months backward in time calculations due to particle stranding (associated with the length of the backward process) and the seasonal variability. Therefore, this normalized color scale is adjusted to highlight the gradient between low and high presence of simulated MP, for each case, as keeping the number of particles counted for each location does not have any signification as it is. Values close to 1 are then the locations for which the studied particles are most likely to come from, where values close to 0 have a low probability of origin and finally blank zones mean that absolutely no particles tracked back in time from the boxes are originating from these areas.

# *3.4.1. Focus on the NB zone*

The tracking results of particles that were statistically found in the fall season in the NB region defined by the red box is shown in Figure 3. This red box corresponds to the area sampled by the i*n-situ* campaign referred to NB in table 1. The tracking has been performed back in time for 3 months, 6 months and one year.

It appeared clearly that 3 months was a retention time very significant for the area as most of the particles found in the fall season seemed to have been already there for 3 months. Then, when looking at the 6 months back-tracking, the results were similar but an extra region, namely the Gulf of Lion (GOL) and the NC path, appeared as another statistical origin of the particles found in Fall in the NB box. Then for the one year back-tracking, as expected, the origin area was more extended to most of the North-Western Mediterranean zone. These numerical results are then in good agreement with the evidenced hot spot of MP concentration in the area, but it also provides two major new information: first, this concentration process seems to have a statistical lifetime of 3 months and second the GOL and the NC path are the main feeders of this hot spot. This result is in good agreement with [Mansui et al., 2015], for which an average retention process of 3 months was highlighted for a few areas, including the NB zone. In addition, the identification of the likely longrange origin is in accordance with the known dynamics of the North-Western Mediterranean basin ([Salat, 1995], [Millot, 1999]), mostly driven by the NC.



*Figure 3: normalized estimation of particles originating from the red box (here NB region), going back in time 3 months (top figure), 6 months (middle figure) or one year (bottom figure). The color scale is the number of counted particles divided by the maximum value for each case.*

# *3.4.2. Focus on the BF zone*

 The tracking results of particles that were statistically found in the fall season in the BF region defined by the red box is shown in Figure 4. The results showed that this target box had for origin a wider area compared to the NB zone (see the previous section). In fall, the particles collected in the south of the box have been there for 3 months whereas those located in the north of

the box found their origin in the GOL mostly and the eastern part of the NC path. For the 6 months back-tracking, a very different result appeared as the origin was not in the box anymore but in a wide area covering the entire NC track including the NB zone and even south of the Balearic Islands. This new area, South of the islands, is an important new information as the particles from this zone partly originated from the incoming Atlantic Waters at the Gibraltar Strait, hence having a completely different origin. More precisely, when looking at the dynamics of the basin, it can be inferred that these particles may be originated either from the North coast of Morocco and Algeria or even from the Atlantic basin, hence, having a widely spread origin. Then, when focusing on the one year back-tracking, it appeared that the particles originated mostly from the North-western part of the Mediterranean Sea, with an emphasis on the NC Eastern track (along the Italian Ligurian coasts and Cote d'Azur French coast).

When splitting this box in two, i.e. a south and a north part, the results supported the fact that for the 3 months reverse in time tracking, the origin was quite different (Figure 5). We observed that for the northern part of the frontal zone the origin was relatively coherent compared to what has been shown so far, with likely origins found along the NC path and the GOL area. However, the southern part of the front (bottom figure 5) showed that most of the likely origin was the red box itself during the last 3 months, even if there was still a certain influence of the GOL and the NC but with a lower impact compared to the northern part of the front. This highlighted the fact that, within the BF, the North part was globally more fed with the north-western Mediterranean circulation when the south part was more subjected to retention.

As suspected for the one-year case in figure 4, if the reverse tracking was made for one year for the south part of the BF, it did not reveal a dominant origin coming from the southern basin, as shown in figure 6 (bottom panel). Atlantic waters flowing through the Gibraltar Strait and feeding this area in MP was unlikely. But if the back in time tracking was made for 6 months (top panel, figure 6), the results showed that the likely origin may be Algerian basin. This result is quite puzzling but at least revealed the strong seasonal variability of the area, partly due to strong wind regimes as discussed by [Seyfried et al., 2019]. This outcome is also supported by [Barral et al., 2021], for which they have shown a very high variability of the number of frontal zones as well as their locations.



*Figure 4: normalized estimation of particles originating from the red box (here BF region), going back in time 3 months (top figure), 6 months (middle figure) or one year (bottom figure). The color scale is the number of counted particles divided by the maximum value for each case.*



*Figure 5: normalized estimation of particles originating from the red box, going back in time 3 months. The color scale is the number of counted particles divided by the maximum value for each case. Top: north of the original red box (BF area) in figure 4; bottom: south part of the original red box (BF area) in figure 4.* 



*Figure 6: normalized estimation of particles originating from the red box (here the south part of the BF area), going back in time 6 months (top figure), and 12 months (bottom figure). The color scale is the number of counted particles divided by the maximum value for each case.*

### **4. Conclusion**

First, from the collected samples, it appears that there is a large spatio-temporal variability in the MP surface distribution, as replicated stations did not give similar results for only 15 days inbetween the measurements. However, every Manta trawl revealed, as expected, MP concentration at the sea surface in orders varying between tens to hundreds of thousands of pieces per square kilometers. The MP concentrations found are in good agreement with previous in-situ MP collection campaigns in the Mediterranean Sea ([De Haan et al., 2019], [Ruiz-Orejon et al., 2018]). The two selected areas, NB north of Mallorca islands and BF the Balearic Front located east of Menorca, showed however different patterns of MP distribution: the NB zone appears to act more as a retention area, whereas the BF zone seemed highly variable and with a diminished retention capacity.

Then, our statistical numerical analysis helped to understand the origin of the MP in these 2 areas. It has confirmed the non-stationary tendency of the MP accumulation in both areas. However, a difference emerged clearly between the 2 zones. The NB zone presented permanent high concentration rates but with a high local variability, as showed by the in-situ measurements, with an average 3 months period of pseudo-stable concentration in the global NB zone as demonstrated by the numerical work. But again, inside this NB zone, local concentrations maximum varied for tens

of kilometers in a couple of weeks, due to the strong ocean currents and weather conditions variability. When tracking backward in time for periods longer than 3 months, the likely origin of this area was the GOL and the NC path, acting as a carrier of MP from the Ligurian and French Riviera coast lines. For the BF area, the concentration rate remained lower that the NB area. The attempt to differentiate the MP origin between the North and the South of the BF area showed that the south part was probably more efficient for retention than the north one. The south part showed a 3-month retention quite important where the north part showed 3-month origins widely spread along the NC path and the GOL. However, due to the high variability of the fronts in this area, the south part also showed important likely origins in the North of the basin. Such results confirmed that the BF is probably not a very efficient retention zone. In addition, it worth noting that all the numerical results are limited by the horizontal resolution of the ocean model used to feed the Lagrangian model. This could be of particular importance when the frontal zones are studied, for which sub-mesoscale dynamics, not solved by the model, could be dominant for MP transport processes. Indeed, higher resolution models, if available, could be used in future studies to compare with the present results to see if retention dynamics is significantly enhanced when sub-mesoscale dynamics is available.

The present study also fairly asks the questions whether there could be some other periods during the year, for which the frontal zones will locally act as efficient retention zones. Our in-situ data did not show that, but our campaign was only during one season (fall) and for only 16 days. The numerical work for the other seasons could be performed, but they would need in-situ confirmation.

Our work also raises the question of a possible transfer from the north part of the BF, where MP did not accumulate much and originate from the NC path and GOL, to the south part, for which accumulation seemed to occur for longer periods, at least during the fall season. Only repeated stations throughout a complete year could answer these interrogations.

In addition, it appears that the overall numerical method exposed here could be used for any ocean region, as long as an ocean general circulation model of sufficient horizontal resolution is used and ensemble lagrangian experiments similar to the presented one are performed. However, whatever the results, the missing information will always come from the lack of MP in-situ sampling replications. It is also worth noting that, at least for this studied area of Mediterranean Sea, replicating the sampling at the exact same locations with a boat cannot guarantee to find concentration of similar orders as proven by our campaign for which the highest densities locations have move 50 km north in 13 days. Among other things, this shows that in-situ campaigns, while remaining of major importance, stay fragile in terms of statistics for regions featuring high mesoscale dynamics.

Lastly, in order to strengthen this latter point, Finite Size Lyapunov Exponents (FSLE) from AVISO products have been calculated for each short period of sampling (e.g stations 2 to 4, or stations 15 to 17). As the results are difficult to interpret, they are not shown here but are available in the supplementary data and we provide here insights of this diagnostic. According to the FSLE maps obtained, most of the stations were located along filaments so one could expect high MP concentrations along these structures as discussed in numerous studies at least for biological materials and supposed for other passive materials [Van Sebille et al., 2020]. But in fact the *in-situ*  data vary with such a wide magnitude that it this difficult to understand whether the sampled areas were isolated, small-scale short-time accumulation zones or areas fed by larger neighboring

accumulation structures. Additional investigations with such similar tools might help to better understand these spatial and temporal multi-scale processes.

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