

Supplementary information for:

## Mineral and biological ice-nucleating particles above the South East of the British Isles

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### Data availability

The data associated with this publication is available from University of Leeds at <https://doi.org/10.5518/979>.

### S1 Surface area distribution through the droplet population

Here we show the results of the Monte Carlo simulations we performed in order to evaluate the distribution of surface area within the droplets of our experiments. We have conducted this simulation for one of the samples with the smallest number of aerosol particles on it, (C022\_1). The simulation is carried out by randomly spreading the particles sampled in each case (from the PCASP-CDP size-distributions) through the droplet population. We assume that the filter inlet system spreads the aerosol particles homogeneously through the surface of the filter, which was tested in a previous study {Sanchez-Marroquin, 2019 #598}. A significant part of the surface area of the samples is constituted by carbonaceous particles of  $\sim 0.1 \mu\text{m}$ , which are less likely to be INPs. Additionally, the smaller aerosol particles are much more abundant and each of them carries a smaller surface area, so their surface area is much better distributed through the droplet population. Hence, we restrict this analysis to particles above  $1 \mu\text{m}$ , which are the ones that could create a large variability in the surface area present in each droplet. The analysis, shown in Fig S1, demonstrates that there is a very low variability in the supermicron surface area present in each droplet of the experiment. The variation is even smaller for the number of particles per droplet.

This variability in the surface area per droplet is much smaller than the one considered in ref. {Knopf, 2020 #683 }. This means that we would not be able to explain our results using a stochastic approach that accounts for the variability of the surface area per droplet. Therefore, the use of the singular description in this study is justified.

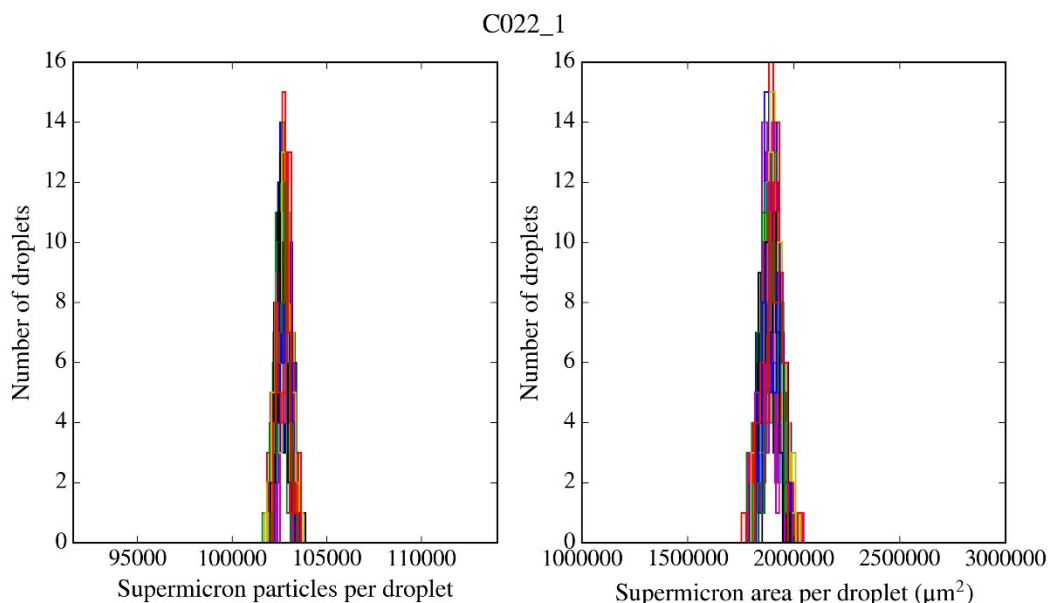


Figure S1. Monte Carlo simulations of the distribution of particles (left) and surface area (right) per droplet of experiment. Simulations have been repeated 45 times and plotted using different colors.

## S2 Drop on and wash off freezing assay comparisons

Some of the samples were analysed in parallel using a similar technique to the one used by a previous study {O'Sullivan, 2018 #469}. We will refer to that technique as “wash-off” droplet freezing assay. This technique consists in suspending the aerosol samples collected on top of polycarbonate filters in 5 mL of pure water (Milli-Q®). This is done by centrifuging the filter (usually only half of the filter in order to save the other half for the SEM-EDS analysis) for about half an hour at ~30 rpm. This suspension is then pipetted onto 22 mm diameter hydrophobic silanised glass slides {Whale, 2015 #293}. Some of the samples have been pipetted on top of a thin layer petroleum jelly {Tobo, 2016 #291}. Before that, we tested that performing the experiment on top of the petroleum jelly surface produced the same results as performing it on top of the glass slide. Then, system is cooled in the same way as the droplet on freezing assay described in Sec. 2.2. The INP concentration of 10 samples was measured using this technique. A comparison in between both techniques is shown in Fig. S2. The agreement is good for the cases a, b, c, g, h and i, while samples d, f and j had some discrepancies within an order of magnitude. However, in sample e, the discrepancy was above an order of magnitude.

The wash-off assay hasn't been performed systematically and its data hasn't been presented in this paper for several reasons. Firstly, this technique produces a much lower INP signal in the experiment than the drop on technique. Hence the technique is not suitable to produce good quality data above the limit of detections on board of the FAAM Bae-146. This is due to the short sampling times that can be normally achieved when sampling on board of a research aircraft. Additionally, we think there were several problems with the aerosol extraction during the course of this campaign, which could explain the low INP signal that this technique produced in some occasions (Fig S2e, i and j). Although we haven't used the data produced

by the wash-off technique, the results produced by this technique and the used drop-on technique are consistent in most cases.

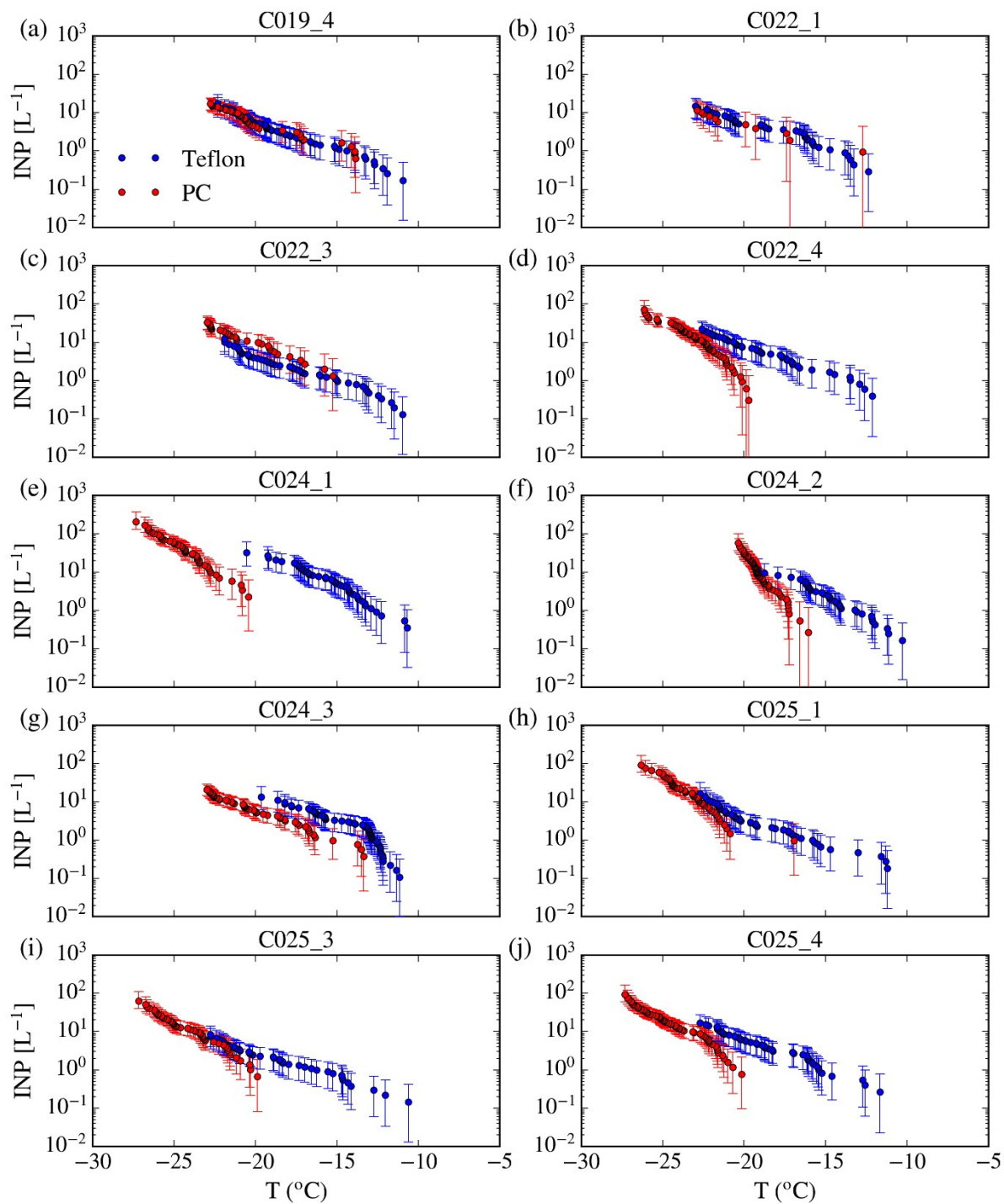


Figure S2. Comparisons between the INP concentration measured by the drop on and wash off technique. Note that the INP signal in the wash-off experiments (Polycarbonate) of samples d, e, h, i and j are only slightly above the limit of detection.

### S3 Correlations

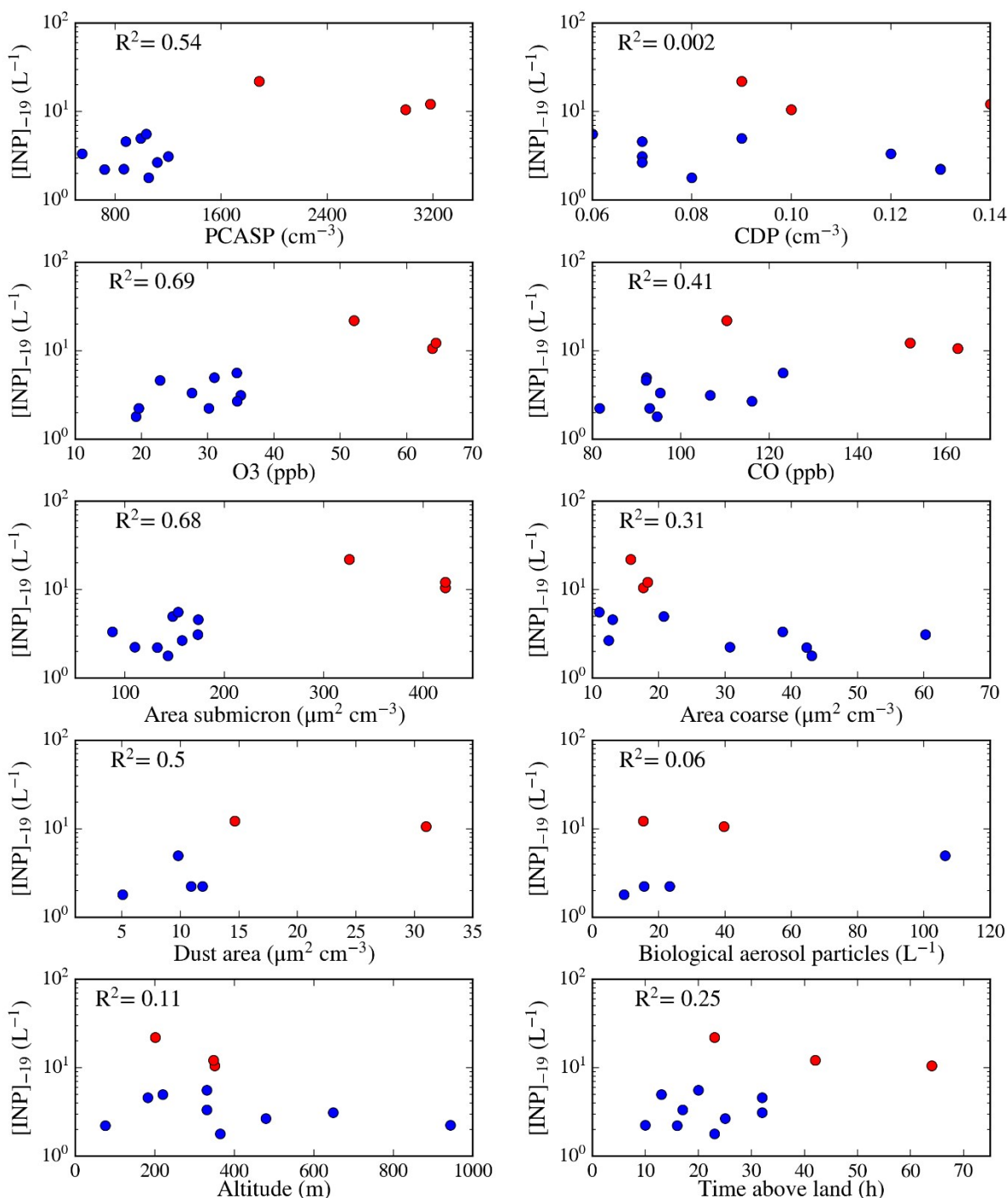


Figure S3. INP concentration at  $-19\text{ }^{\circ}\text{C}$  represented against different variables measured by the BAe-146 such as the number of aerosol below and above  $\sim 3\text{ }\mu\text{m}$  (PCASP and CDP respectively), CO and O<sub>3</sub> concentration, altitude, area of the coarse and submicron modes measured by the PCASP and CDP. The data has been also plotted against surface area of dust and number of biological aerosol particles measured using SEM-EDS, as well as time over land (from the HYSPLIT analysis). Data from the C024 flight is presented in red while the rest of the data is represented in blue.

### S4 Size distributions of the samples

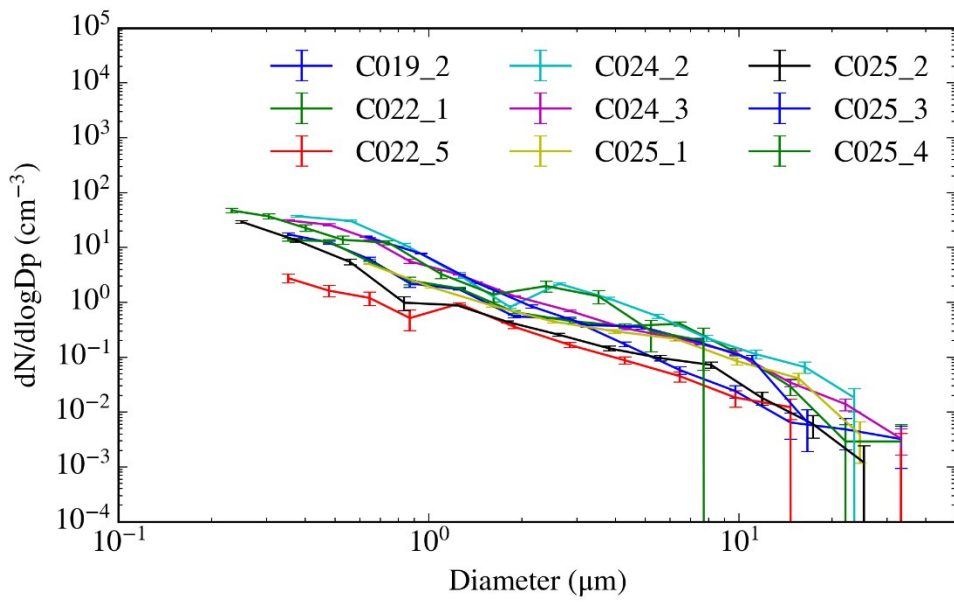
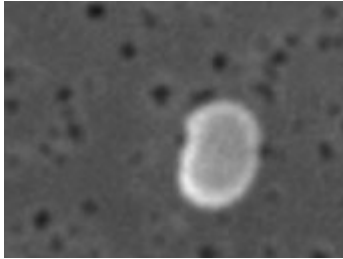


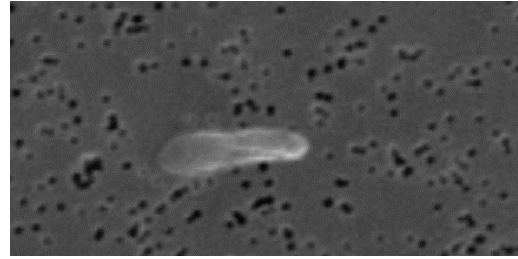
Figure S4. SEM-EDS number size distributions of all the analysed samples. The data shown here have been obtained in the same way as the one shown in Sect 3.2.

## S5 Identification of primary biological aerosol particles

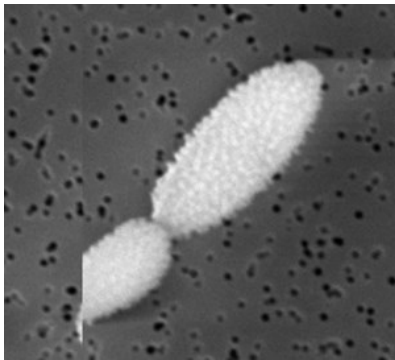
Here we show a selection of ~100 biological aerosol particles detected in our samples with a description of the most likely genus or phylum they belong to. The size can be inferred given that the pores in the filter have a diameter of 0.4  $\mu\text{m}$ .



Unknown spore  $\approx 2\mu\text{m}$   $\varnothing$  possibly *Penicillium* or *Aspergillus*.



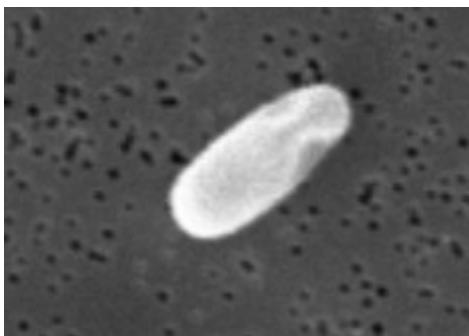
Unknown  $8\mu\text{m}$  ascospore composed of two cells.



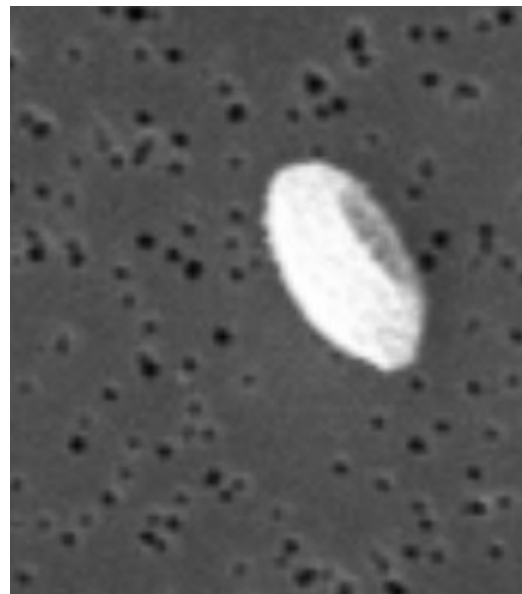
Chain of two *Cladosporium* conidia.



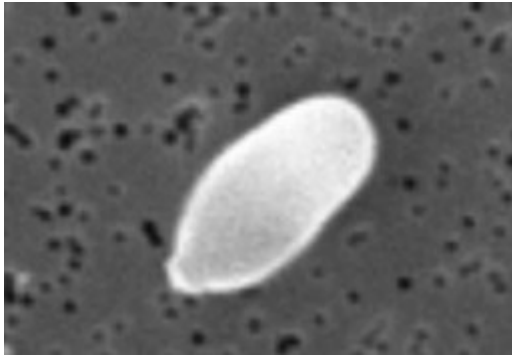
$12\mu\text{m}$  elongated oval – unknown ascospore.



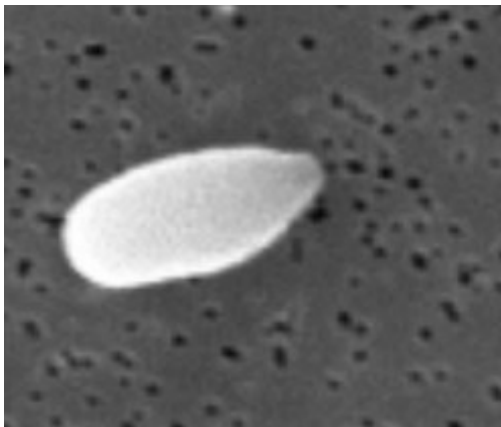
Oval spore with two lobes folded in near end, no attachment scar. Probably an ascospore but unknown species.



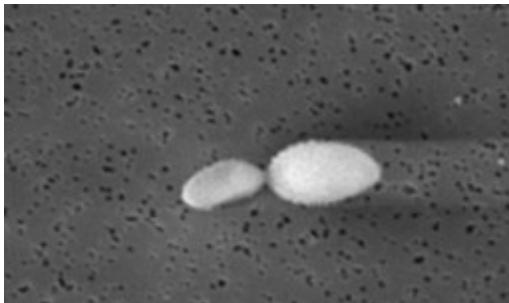
Conidium of *Cladosporium* spp.



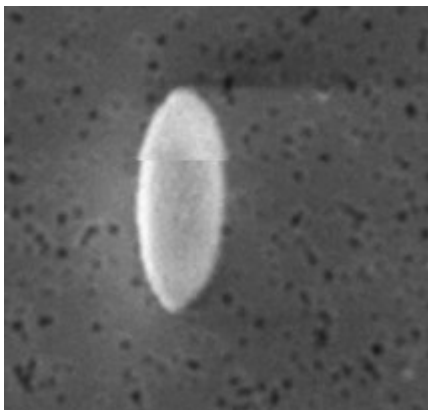
Cladosporium.



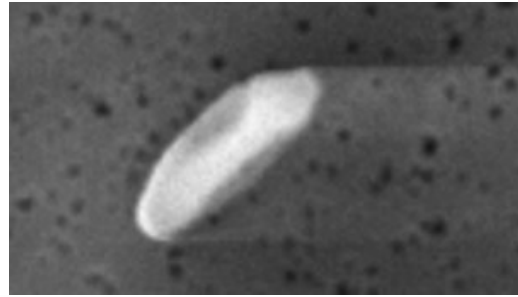
Cladosporium.



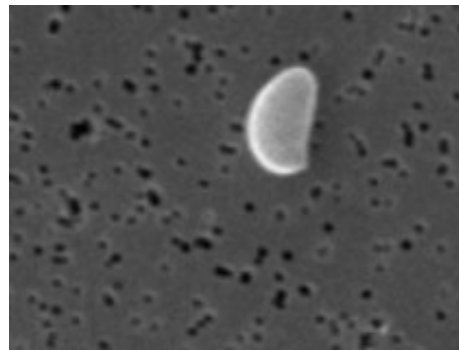
Cladosporium.



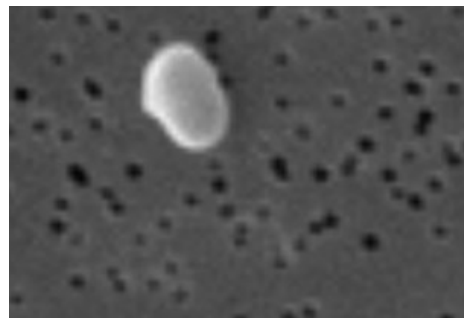
Cladosporium.



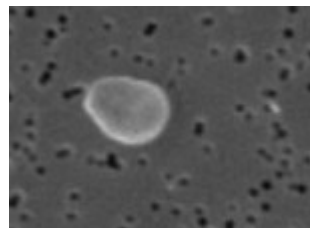
Oval with attachment scar – Unknown conidium (could be smooth type of Cladosporium).



Small size basidiospore (around 3-4  $\mu\text{m}$ ) probably a species of bracket fungus.

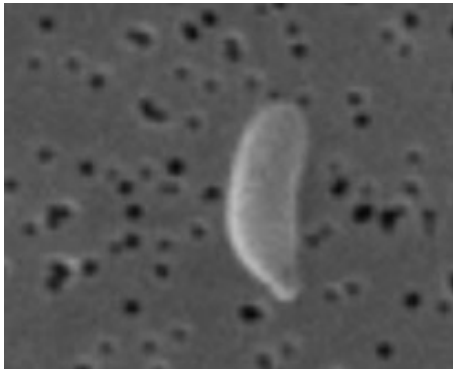


Oval spore but possibly with a ridge 1/3 way down. Unidentified.





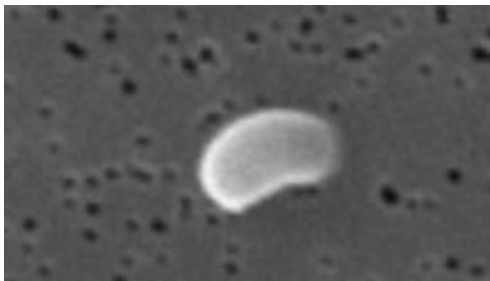
Oval-pear shaped possible constriction in centre – unknown.



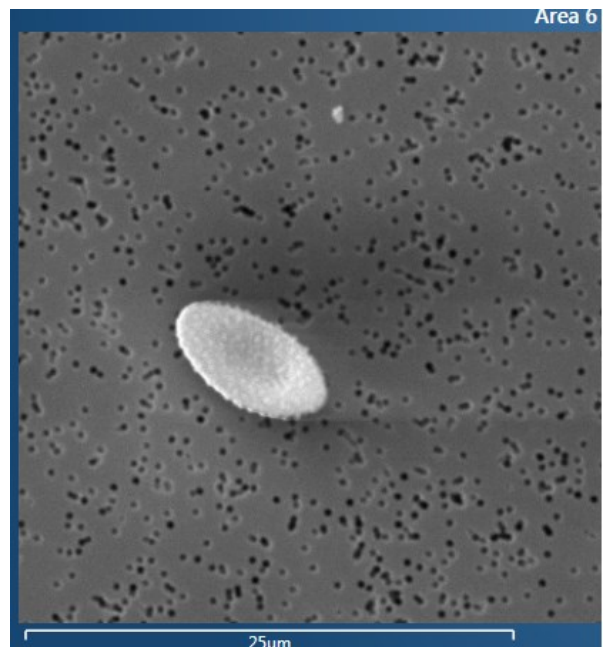
6-7 $\mu$ m Basidiospore – possibly Pleurotus.



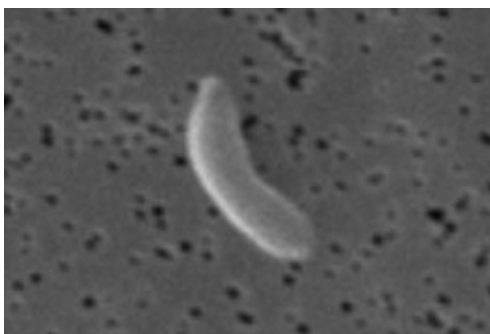
Cladosporium spp.



Kidney-shaped possible septum in centre. Asymmetry suggests likely to be a basidiospore.

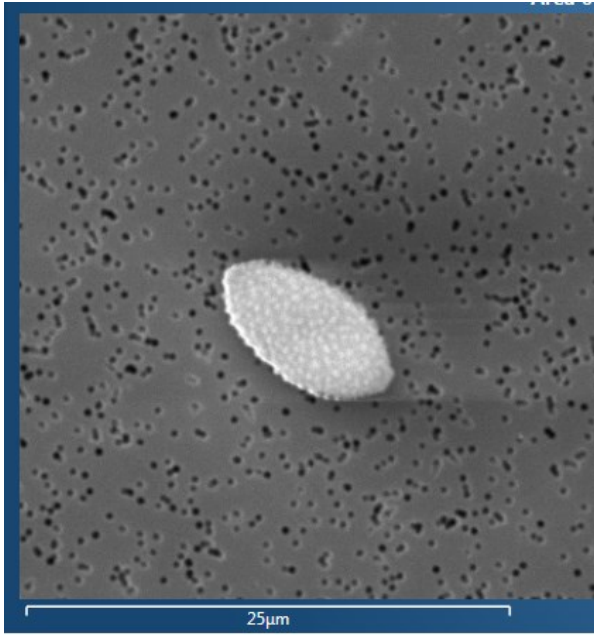


Cladosporium spp.

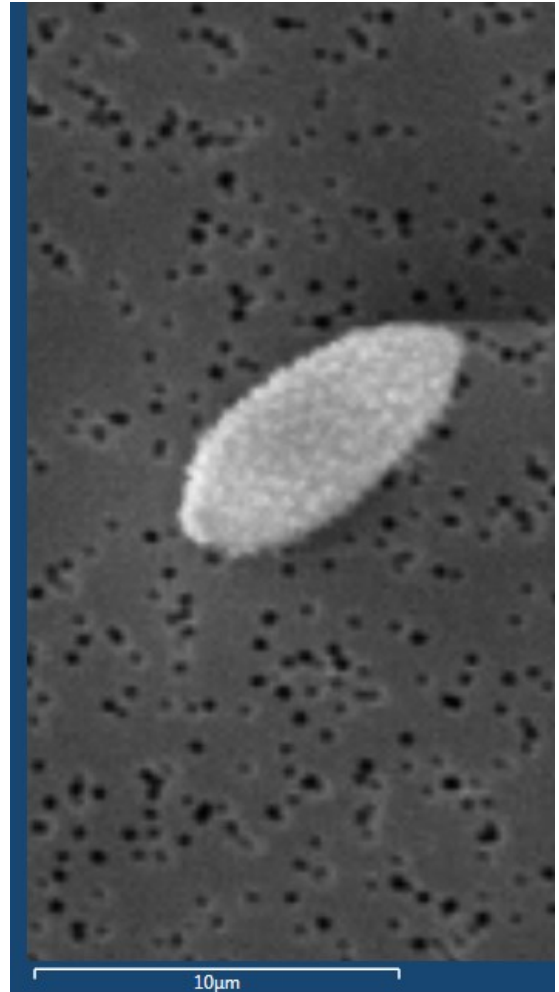


Elongated/curved oval – unidentified fungal spore.





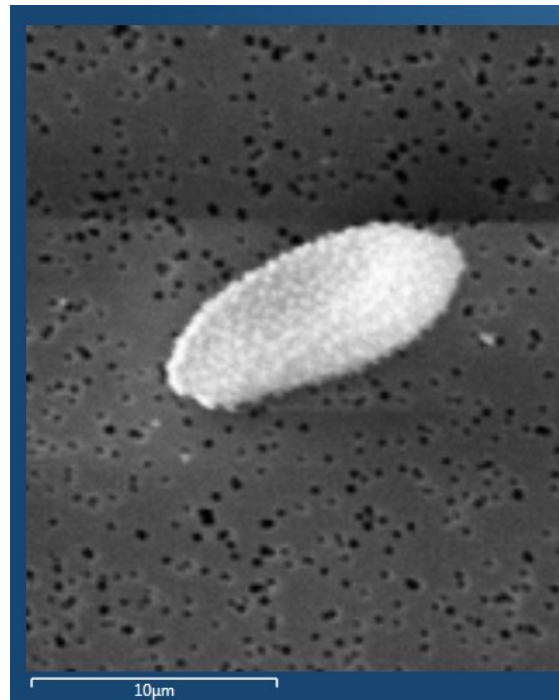
Cladosporium spp.



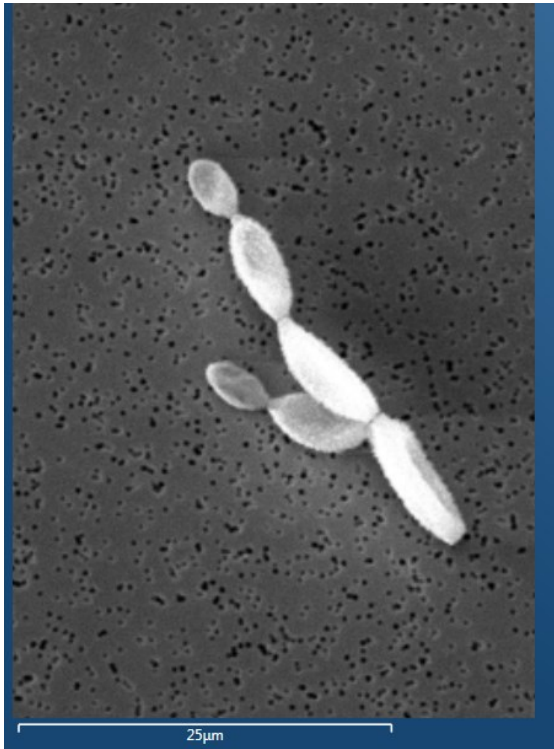
Cladosporium.



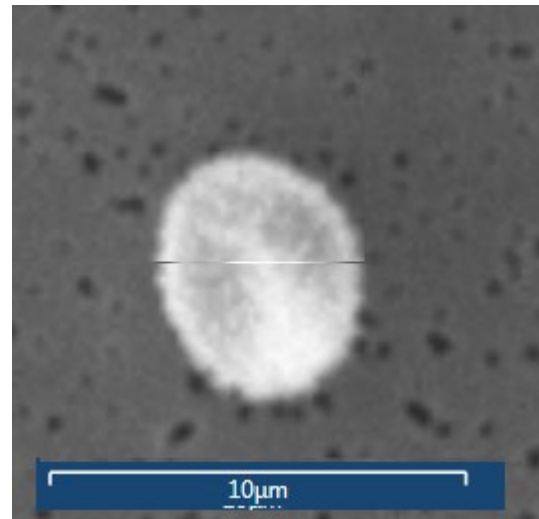
4-celled ascospore slightly dehydrated  
unknown species.



Cladosporium.

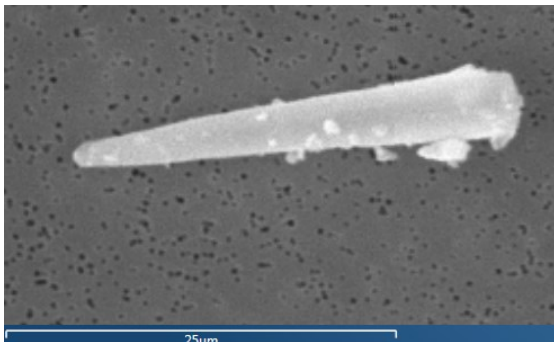


Cladosporium.

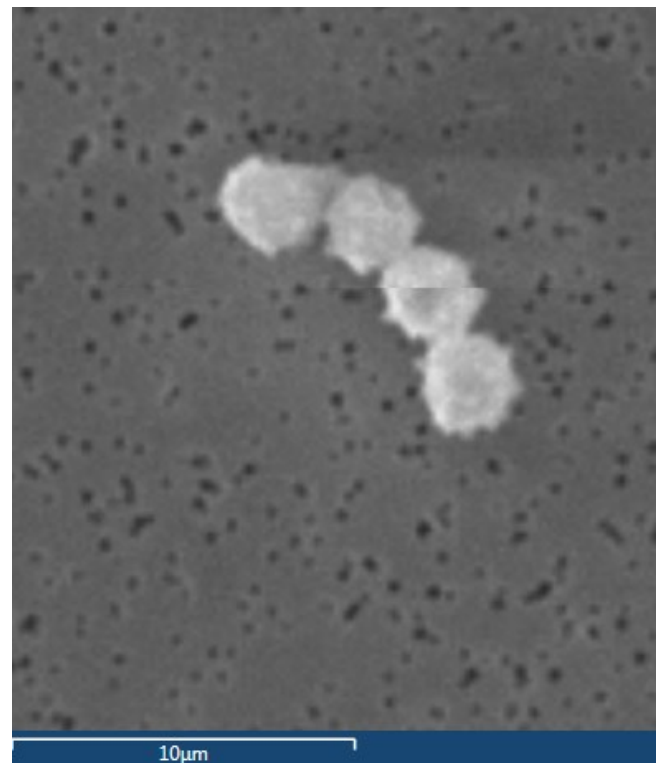
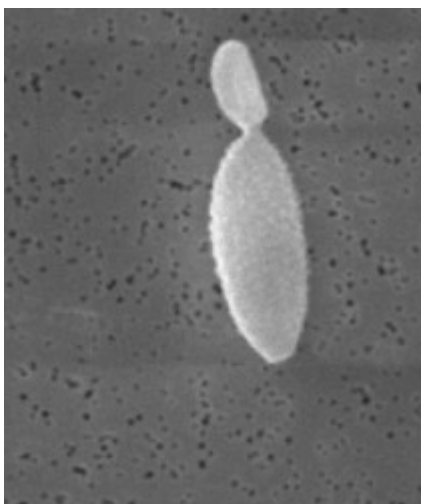


Oval-spherical, rough surface – possibly Chrysosporium or Aspergillus.

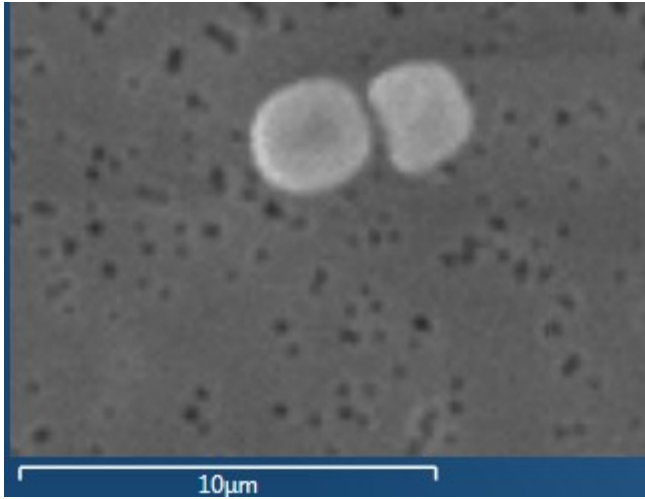
Cladosporium.



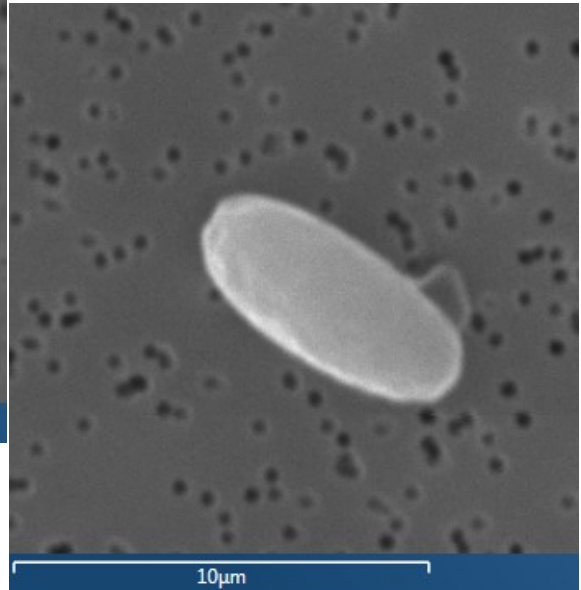
Probably plant hair/spine.



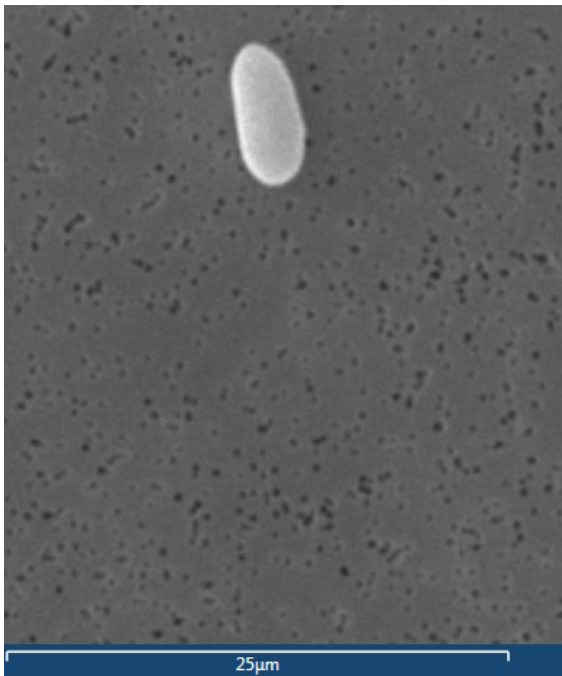
Aspergillus or Penicillium spp.



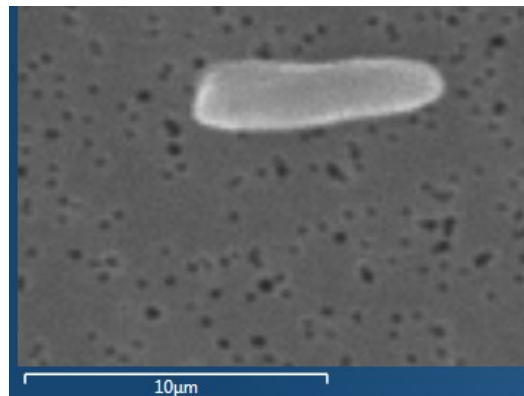
Aspergillus or Penicillium spp.



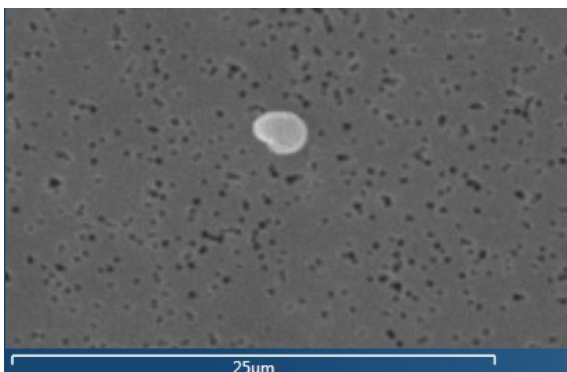
Oval conidium with attachment scar.  
Possibly Botrytis spp or Sporothrichum.



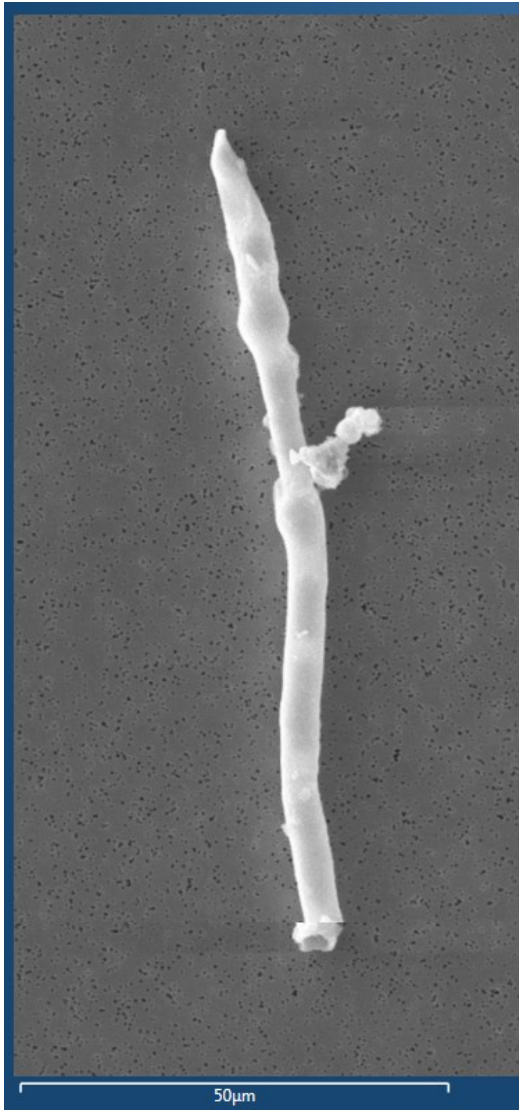
Oval spore – probably an ascospore but unknown.



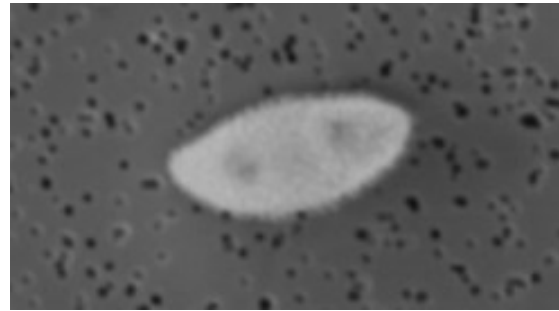
Elongated oval spore, some asymmetry suggests possibly a basidiospore.



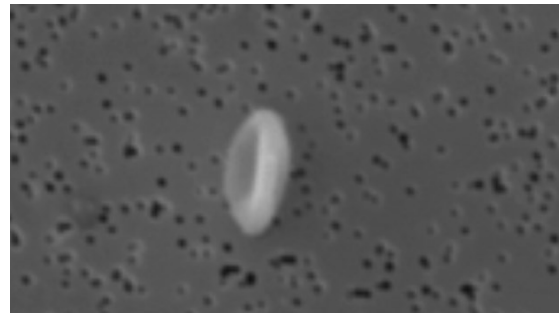
Possible ballistospore.



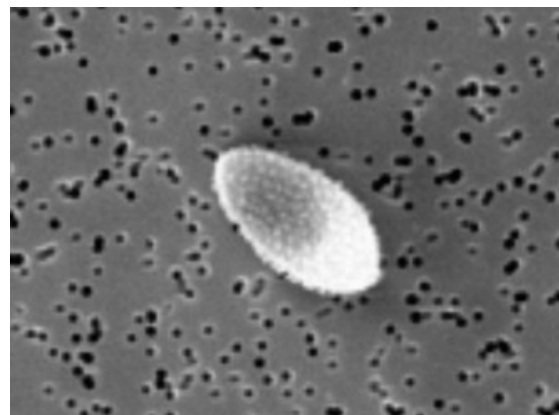
Fungal hyphal fragment with septae visible.



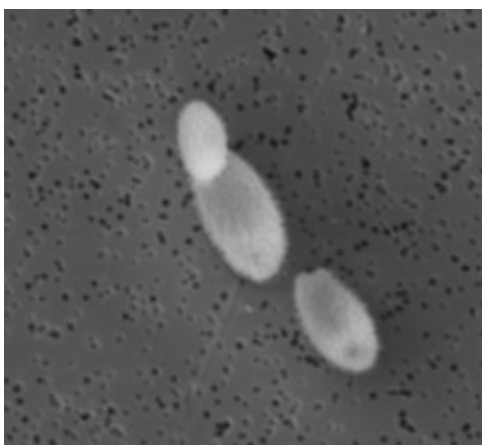
Cladosporium.



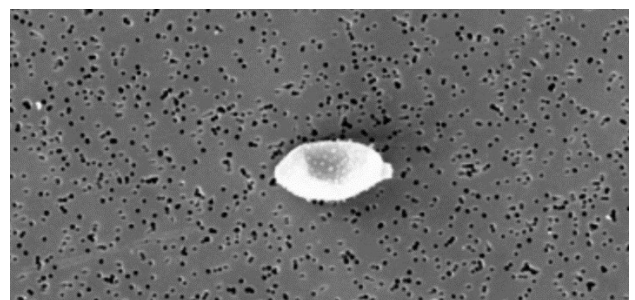
Unidentified.



Cladosporium.

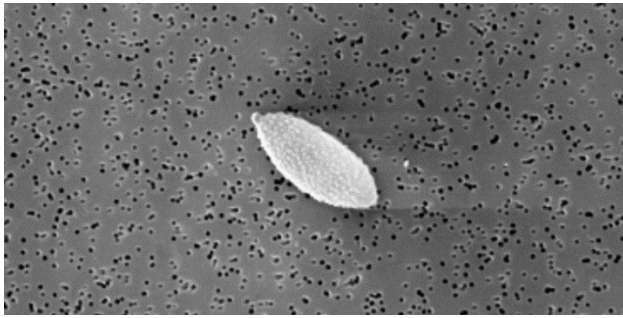


Chain of spores likely broken on impact onto the filter – typical of Cladosporium species.

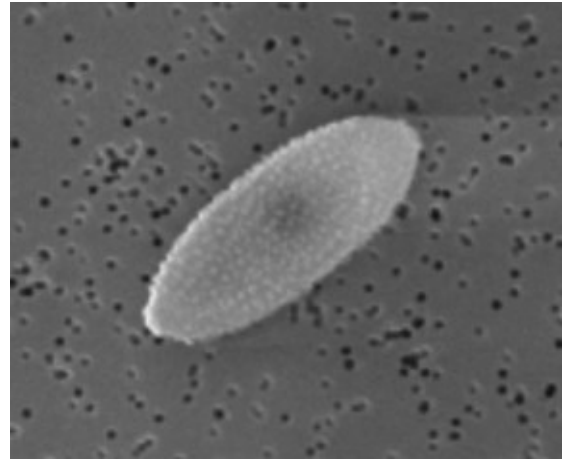


Cladosporium.

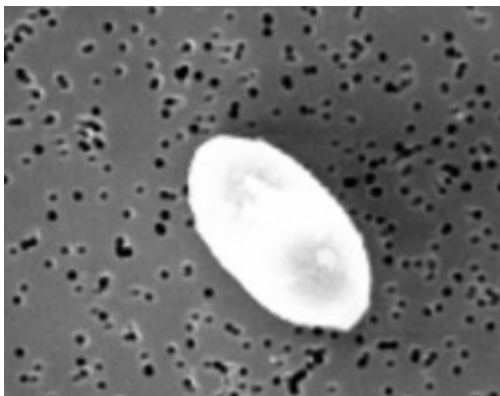




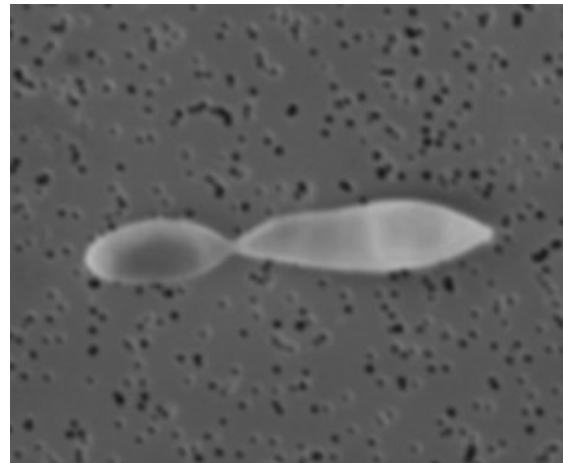
Cladosporium.



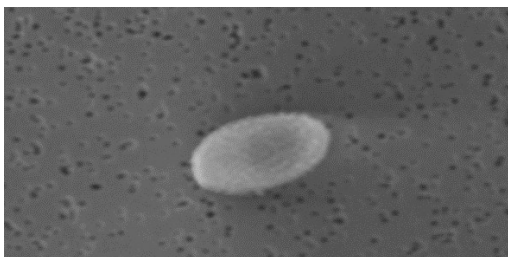
Cladosporium.



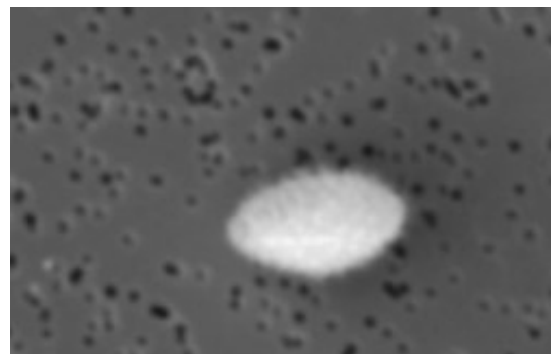
Fungal conidium – possibly Cladosporium.



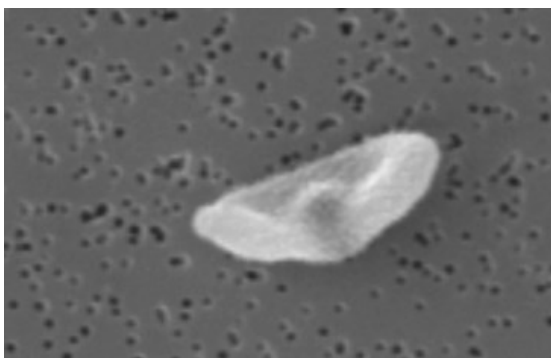
Smooth-type Cladosporium.



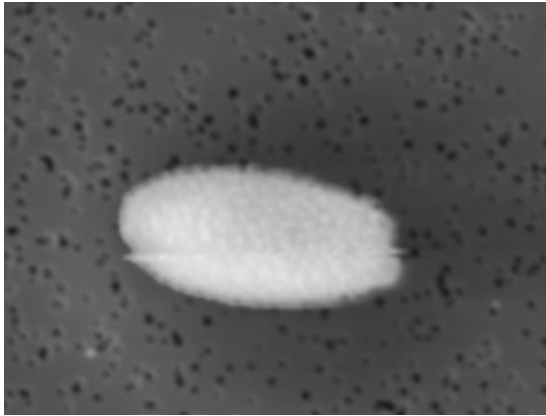
Cladosporium.



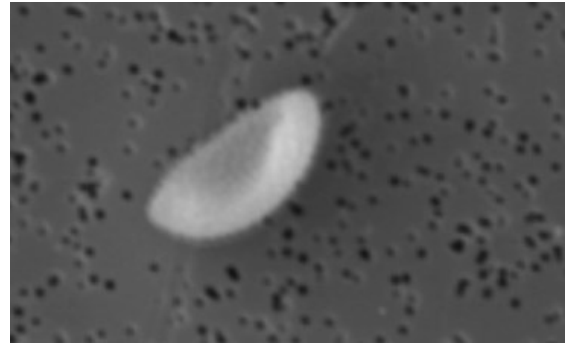
Unknown – possibly Cladosporium.



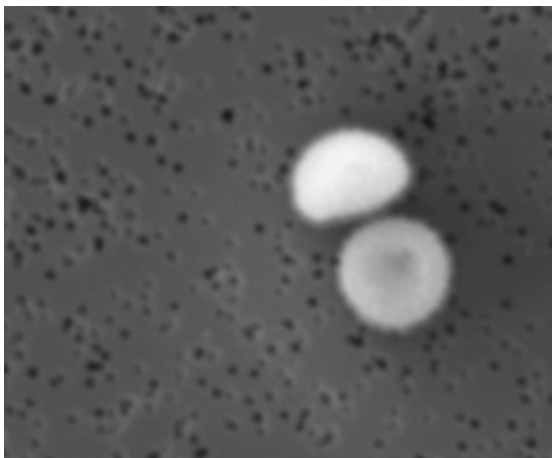
Desiccated Cladosporium.



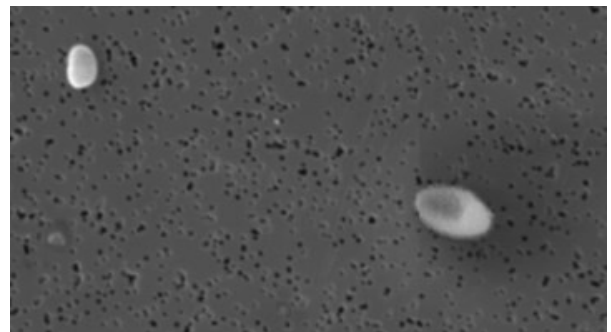
Cladosporium.



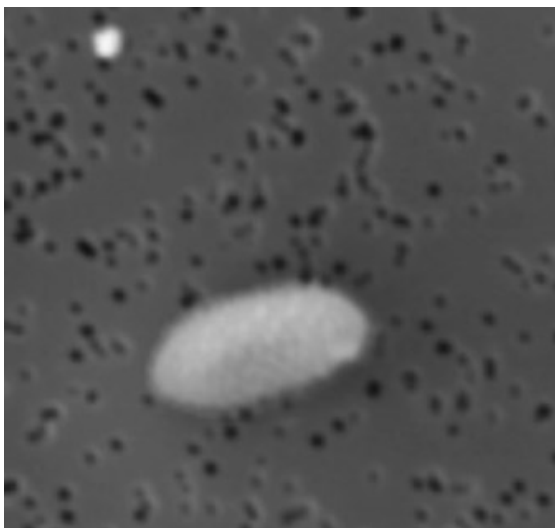
Cladosporium.



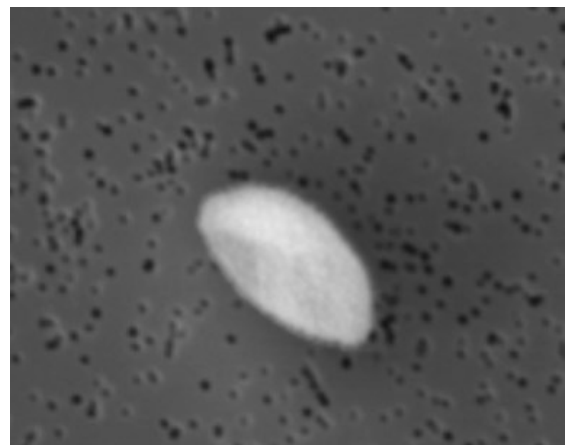
Penicillium or Aspergillus.



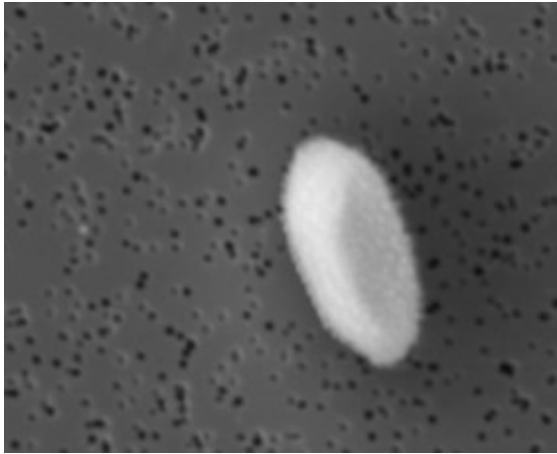
Cladosporium (Right) Penicillium or Aspergillus (left).



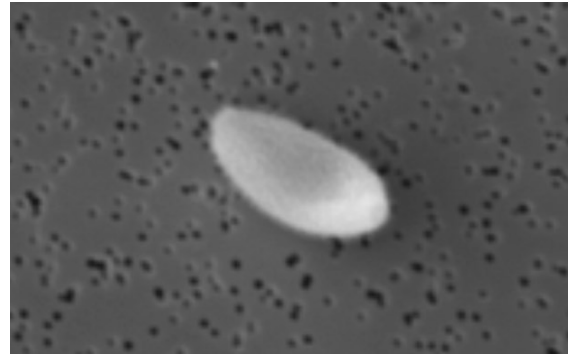
Cladosporium.



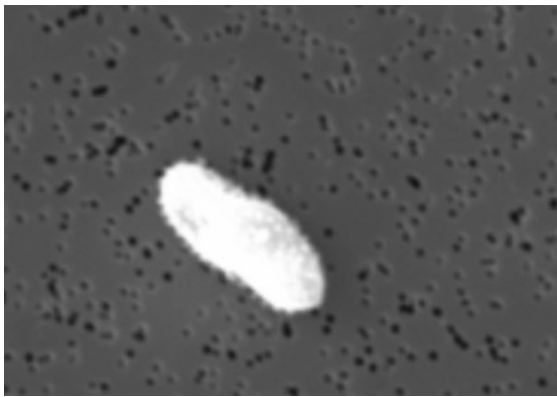
Cladosporium.



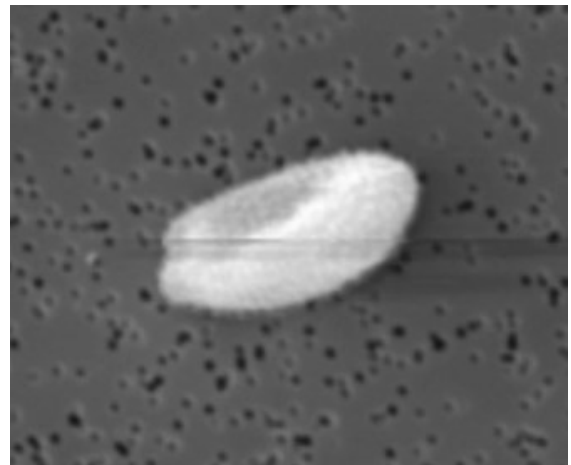
Cladosporium.



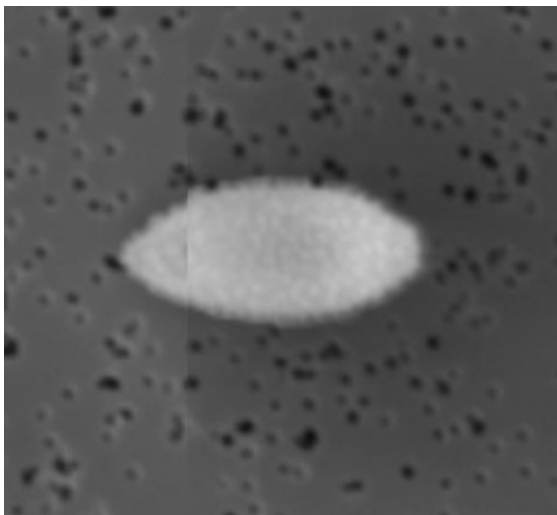
Cladosporium.



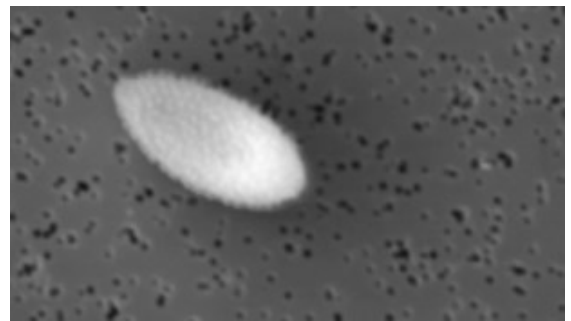
Cladosporium.



Cladosporium.

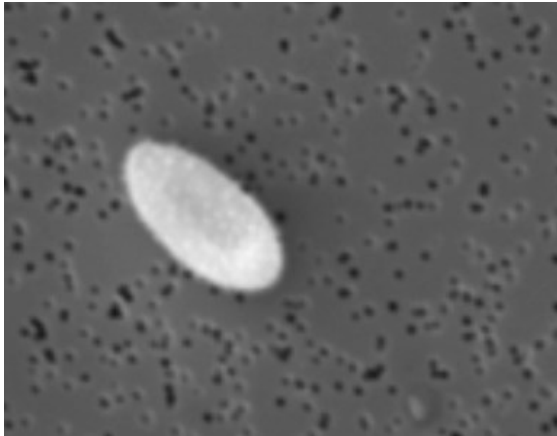


Cladosporium.

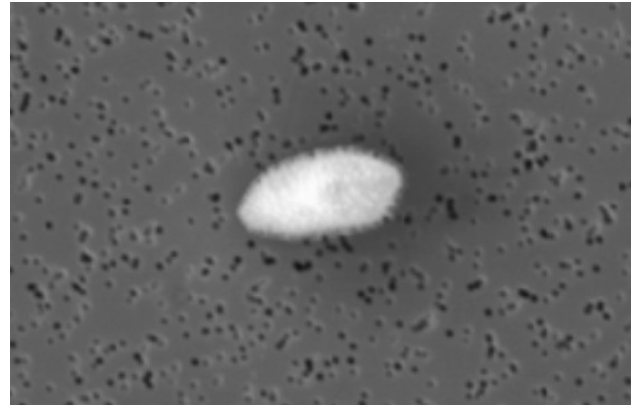


Cladosporium.

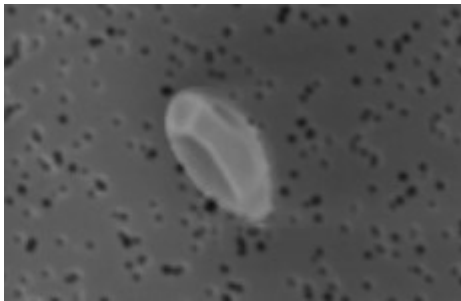




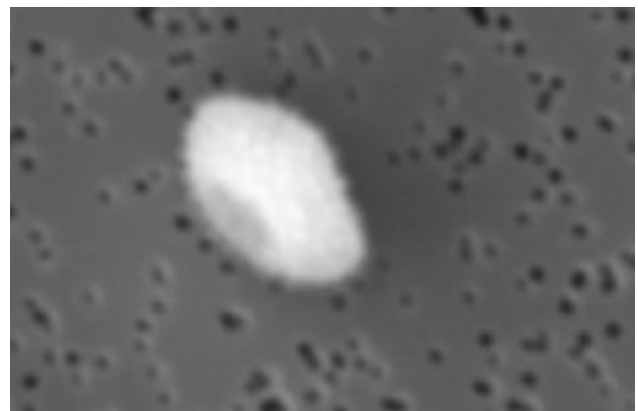
Cladosporium.



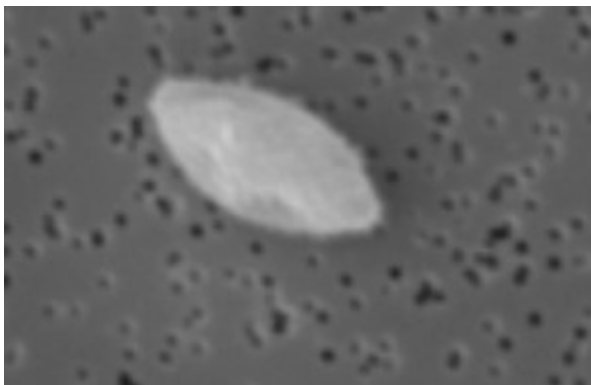
Cladosporium.



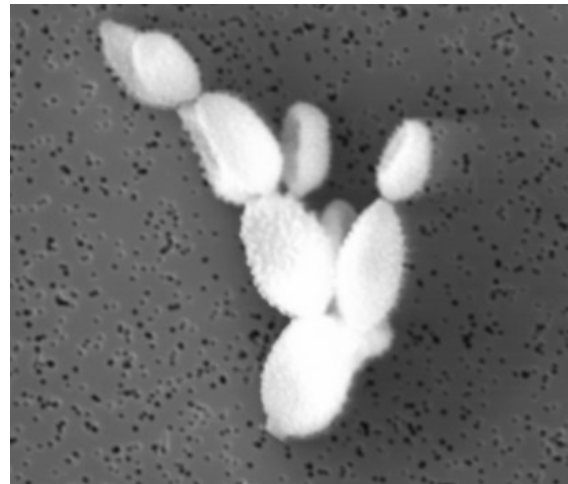
Unknown conidium.



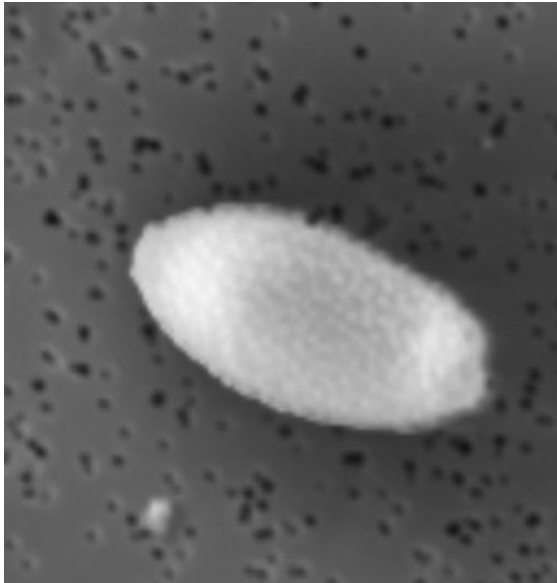
Cladosporium.



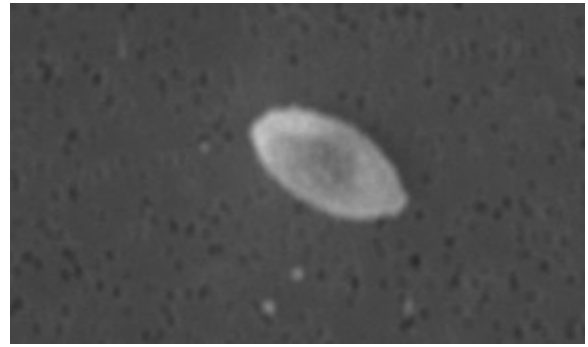
Cladosporium.



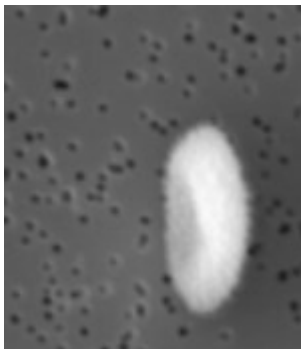
Cladosporium.



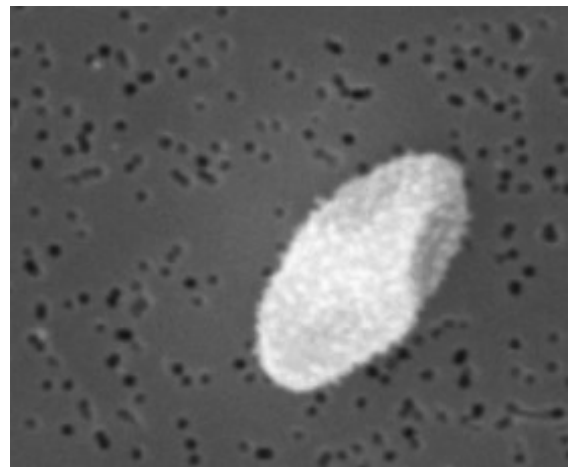
Cladosporium.



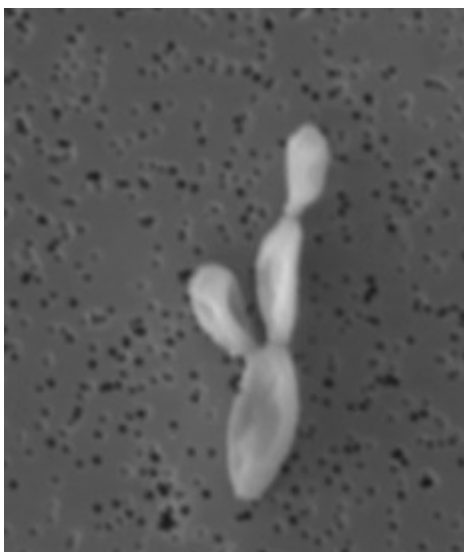
Cladosporium.



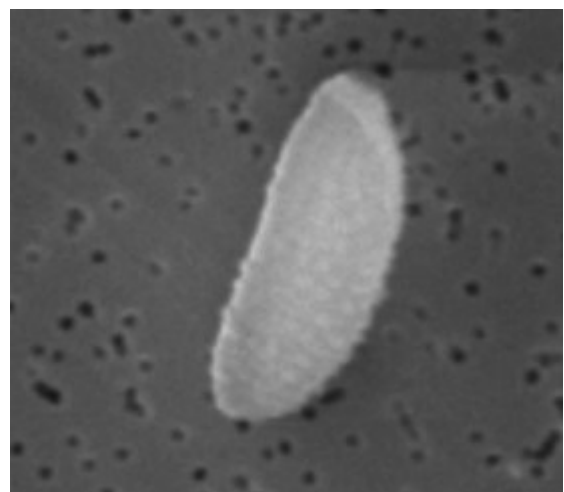
Cladosporium.



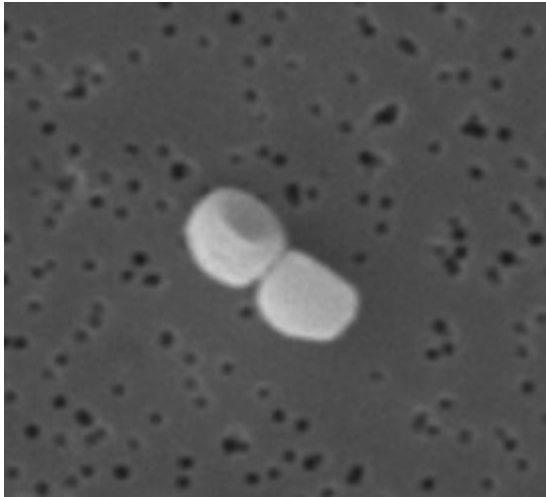
Cladosporium.



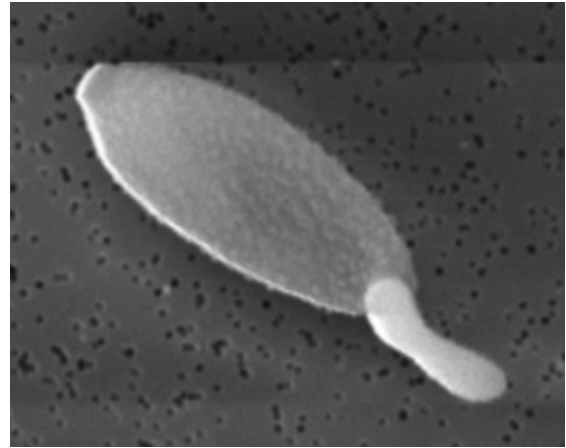
Cladosporium.



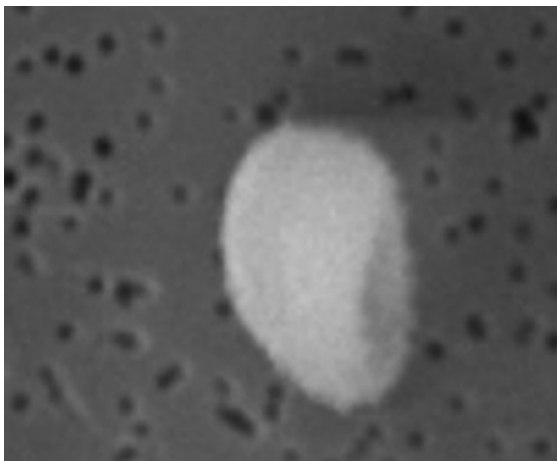
Cladosporium.



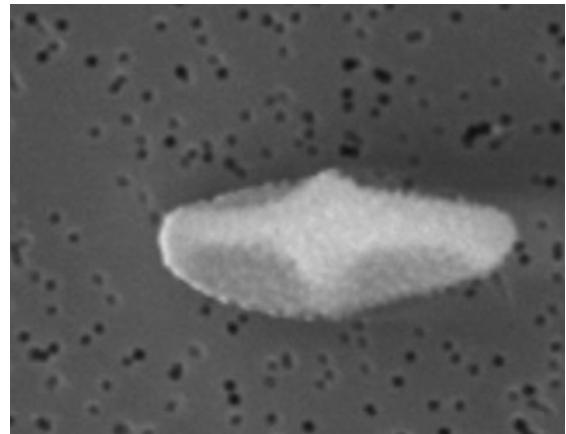
Penicillium or Aspergillus.



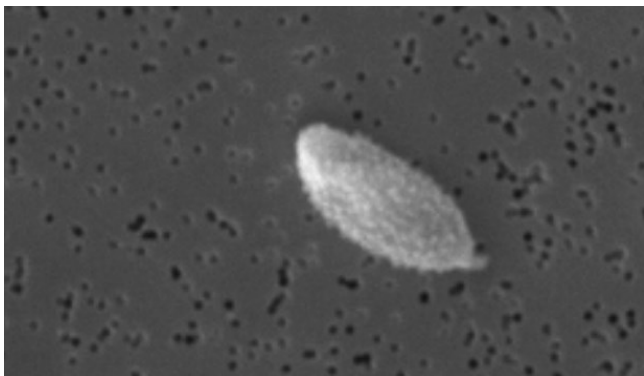
Cladosporium, which appears to be germinating.



Cladosporium.



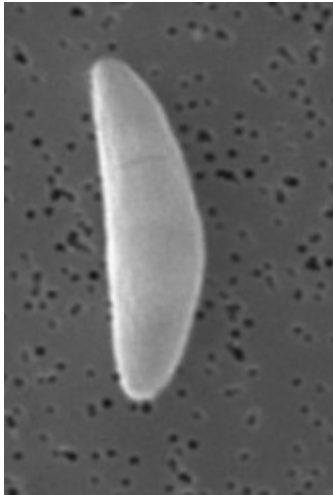
Cladosporium.



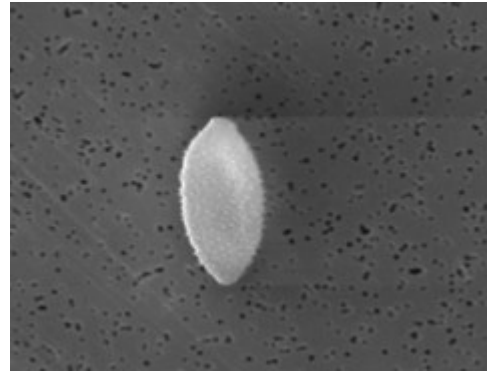
Cladosporium.



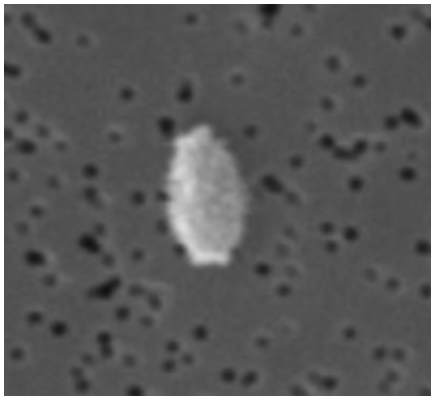
Didymella.



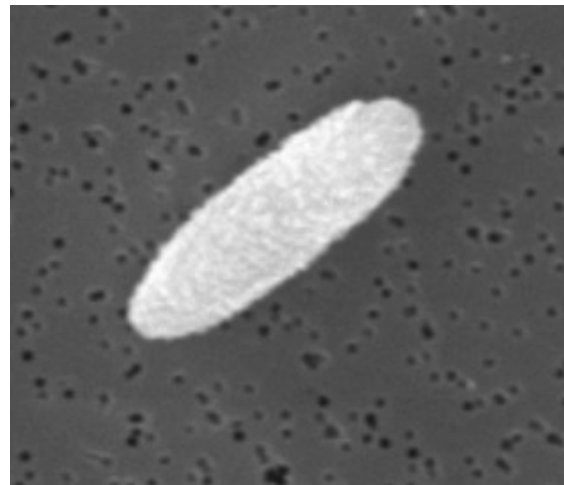
4-celled ascospore.



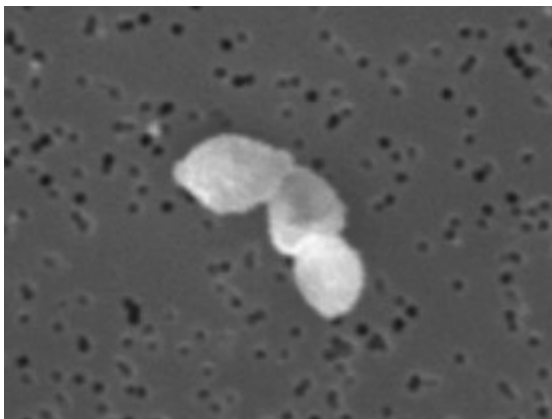
Cladosporium.



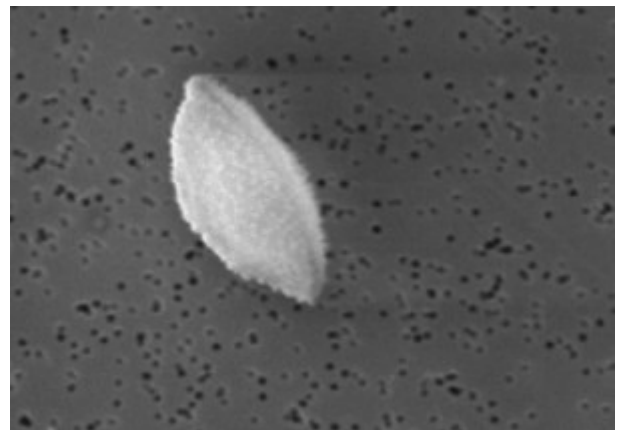
Unknown conidium.



Conidium- possibly Blumeria.



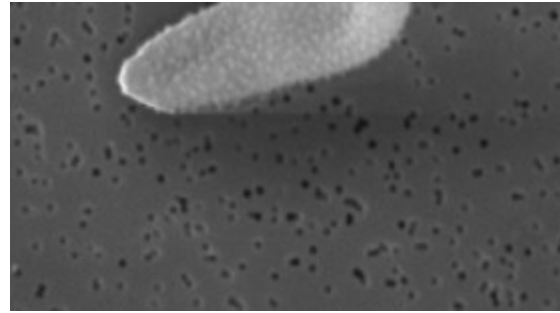
Penicillium or Aspergillus.



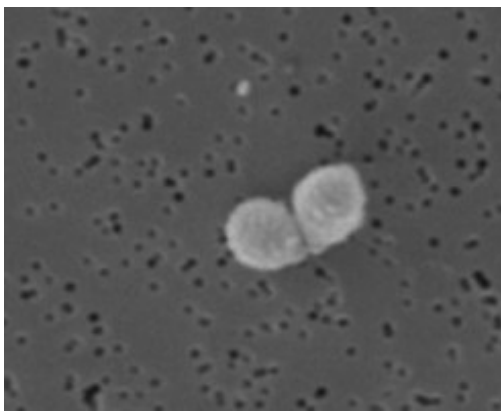
Possible basidiospore but could also be a conidium deformed by desiccation.



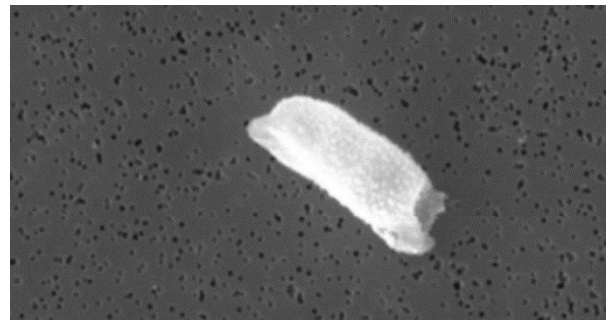
Unknown.



Cladosporium.



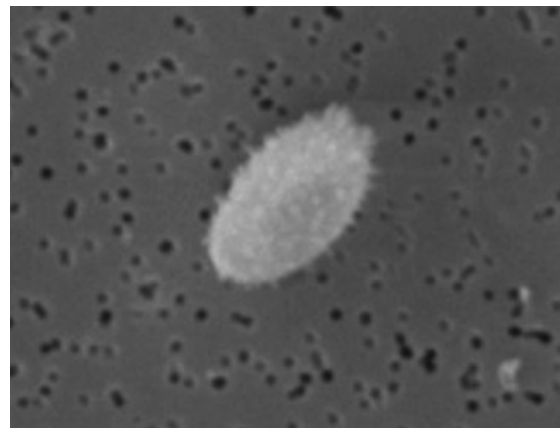
Penicillium or Aspergillus.



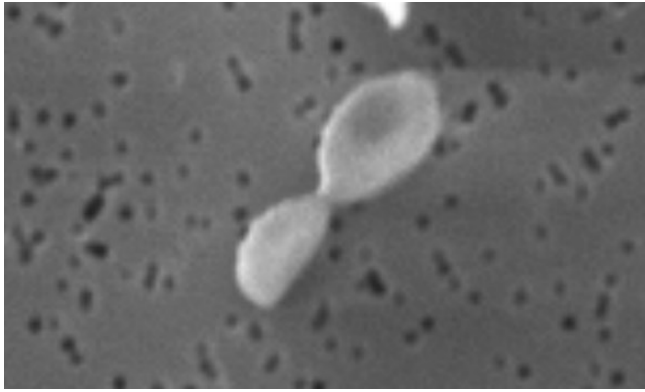
Possible basidiospore but could also be a conidium deformed by desiccation.



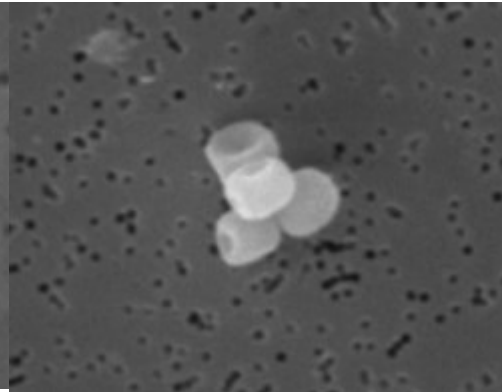
Cladosporium.



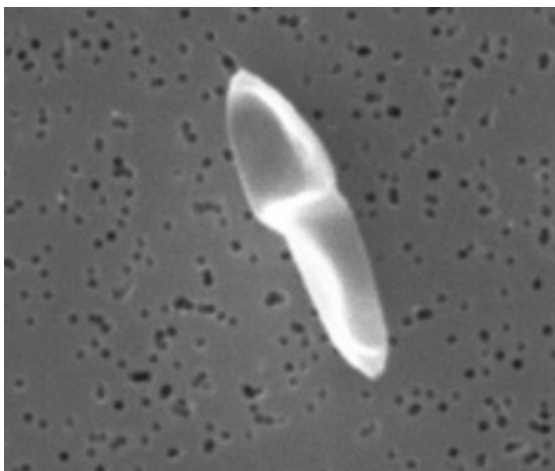
Cladosporium.



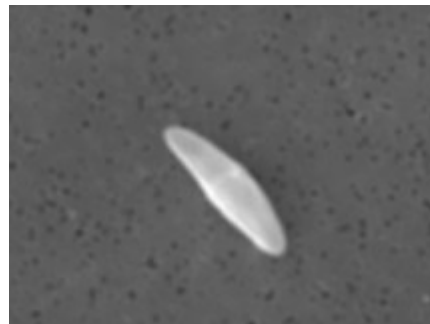
Cladosporium.



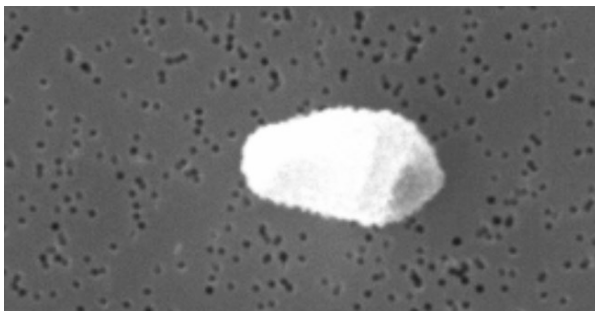
Penicillium, Aspergillus or Wallemia species, shape may be affected by desiccation for SEM imaging.



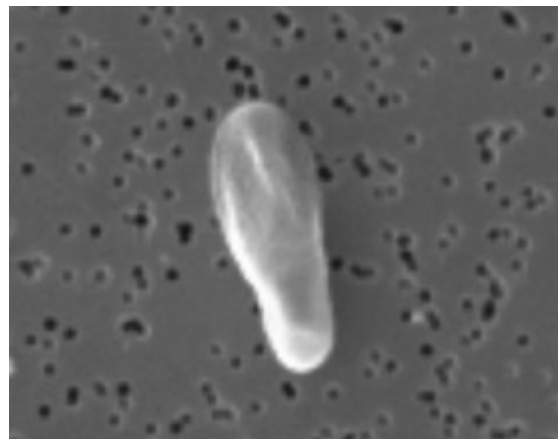
Ascospore, possibly *Mycosphaerella graminicola*.



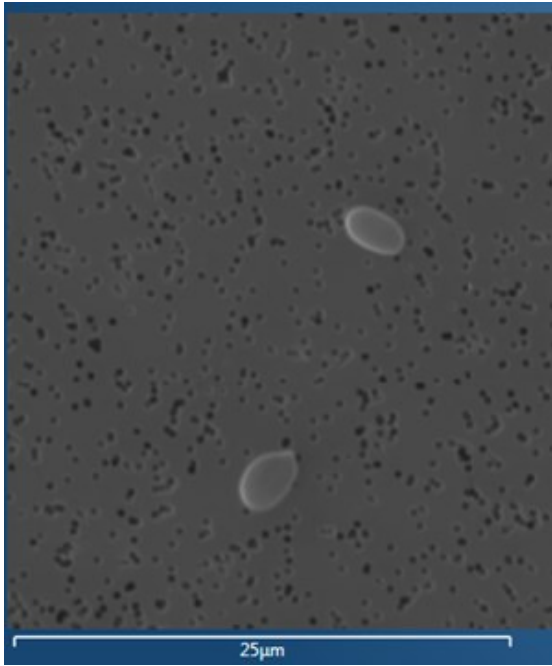
Ascospore – possibly *Didymella* spp.



Cladosporium.

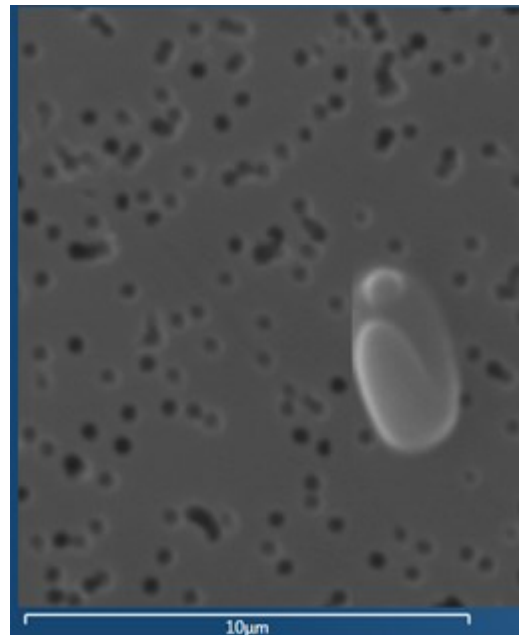


Unidentified ascospore.



~ 3.5um fungal spores – possibly basidiospores or conidia.

Possible basidiospore or conidium.



Possible ballistospore.

