Effects of marine biology and air-mass trajectories on cloud brightness

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Introduction

The cloud droplet number concentration (N_d) of liquid clouds is related to cloud albedo, which is an important factor in the Earth's energy budget (Wood, 2012). The effects of marine aerosol on N_d are still not well characterised in climate models, and thus present an uncertainty in not only the present day climate, but also in our knowledge of pre-industrial climate. Using a combination of satellite data and modelling, this project aims to investigate and characterise the effects of ocean biology and sea spray aerosol on N_d in the pristine Southern Ocean without the influence of anthropogenic aerosol emissions.

Methods

In order to account for all air that ends up in a given cloud over the Southern Ocean, backwards trajectories were simulated using FLEXPART using non-reactive air tracer particles.

MODIS cloud data

The retrieval of N_d from satellite data is possible by using remotely sensed cloud effective radius, cloud optical depth and cloud top temperature (Grosvenor et al., 2018). Other cloud properties, such as liquid and ice cloud effective particle radius, optical thickness and water path, cloud top properties (day and night pressure, temperature, and height) are readily available as MODIS data products (Platnick et al., 2017).

FLEXPART

A forward trajectory analysis of air-mass tracer particles (AIRTRACER) will be carried out using the FLEXPART software (Pisso et al., 2019). FLEXPART is a Lagrangian particle dispersion model that uses 3-hourly global ERA5 ECMFW reanalysis data at 0.5x0.5 grid resolution and 137 pressure levels as input to calculate particle movements. Particle concentrations calculated by FLEXPART are output every time step (10 minutes) onto a 3D grid. The positions of individual particles are also saved at each time step so that the trajectory of each individual particle is known.

AIRTRACER particles are non-reactive and are not scavenged by cloud processes or precipitation, ensuring that particles are able to move to the free troposphere through clouds without being removed from the simulation.

The model was ran with convective atmospheric boundary layer parameterization turned on, improving the accuracy of the dispersion of the tracer particles.

Explanatory variables

Trajectory variables

Possible explanatory factors relating to air-mass trajectories were calculated for each cloud pixel from the FLEXPART data. These included how much time trajectories spent in the boundary layer and the free troposphere in the days leading up to their arrival to a cloud. The boundary layer height we use is defined by the ECMWF data FLEXPART uses as the minimum height for the bulk Richardson number reaching the value of 0.25. Other factors include the mean altitude of trajectories and time spent within 200m of sea surface.

Meteorological variables

Meteorological factors were taken from ERA5 REANALYSIS data and collocated with particle trajectories. These included wind speed, ocean wave speed, total precipitation, sea surface temperature, air temperature, sea surface salinity and sea surface height above geoid.

Biological ocean surface variables

Chlorophyll exposure, depth of the euphotic layer depth, mixed layer depth, sea surface zooplankton concentration were all included in the analysis as tracers of phytoplankton biomass, ocean productivity and DMS. The amount of time air-masses interacted with the ocean surface was also considered to calculate the exposure to these variables, as well as their mean values while air-masses interacted with the ocean surface.

Dataset description

Data filtering

This analysis focuses on clouds that are unaffected by anthropogenic pollution. In this analysis, we define "clean" clouds as clouds whose air-mass trajectories do not cross any coastlines other than Antarctica. This is dependent on how long the trajectories are run for, and at the 9 day mark, none of the cloud pixels in this simulation met this criterion, as shown in figure 1. Previous research (Clarke et al., 1998; Dall'Osto et al., 2017; Fossum et al., 2018; Freney et al., 2021; Hara et al., 2021) has also used 5- or 3-day back-trajectories in order to investigate sea surface aerosol sources. The fraction of clouds unaffected by continental air masses decreased steadily with trajectory length, with only 2% of clouds remaining uninfluenced by continental air masses after 8 days, and no clouds remaining after 9 days. A trajectory length of 5 days was used to define clean clouds, as at this time, 160 cloud pixels (39%) still met the criterion to be considered clean trajectories. The locations of these pixels, and their N_d, are shown in figure 2.

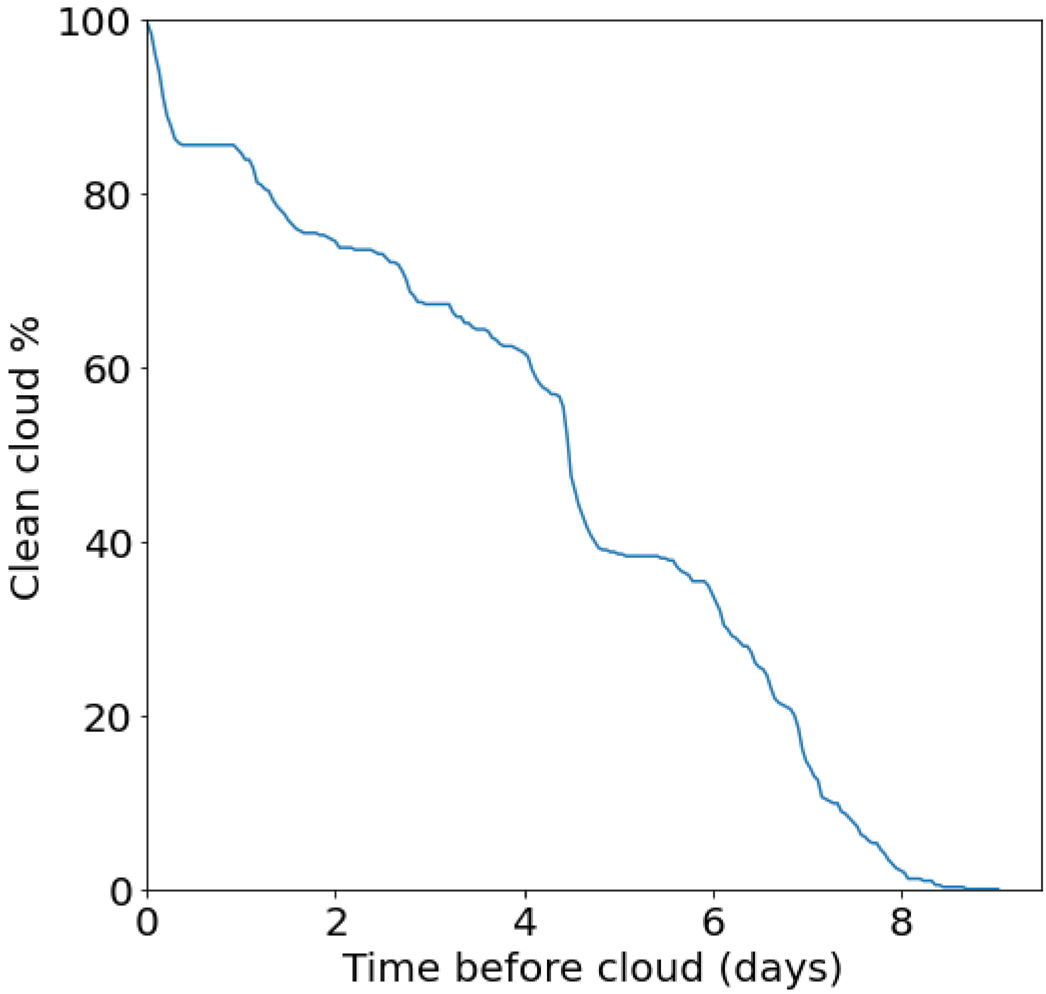

Figure 1. Percentage of cloud pixels considered “clean” (not interacting with any land masses) as a function of trajectory length for the 415 cloud pixels in this analysis.

The study area includes the Kerguelen Islands, which is an ice free land mass and is inhabited by scientists. Although the backward trajectories did not indicate any of the “clean” trajectories being influenced by this island, some cloud pixel locations have been flagged as “possible island influence” based on wind speed and direction at the cloud pixel locations. Satellite images show clearly that clouds downwind of this island have different properties to others in the region. Figure 2b also clearly shows elevated N_d in locations downwind from the island.

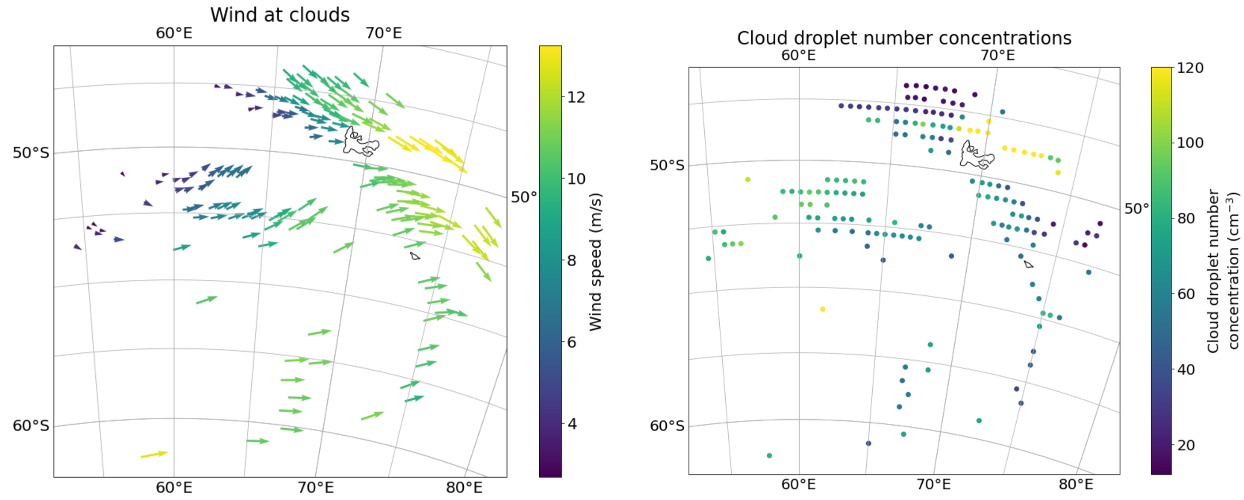

Figure 2. "Clean" cloud pixel locations included in the final dataset. Wind speeds (a) and N_d (cloud droplet number concentrations) (b) at each cloud pixel shown. Cloud pixels downwind of Kerguelen Islands have higher N_d than others.

Correlations with explanatory variables

Correlation coefficients were calculated at 24 hour intervals, with hour 0 being the time air masses are in the clouds. Ocean surface properties are cumulative means over the periods where air masses are in the boundary layer, and averaged over the 250 trajectories for each cloud pixel. Variables that showed a consistent correlation with other variables were removed as duplicates.

For the overall dataset, which includes both group 1 and group 2 cloud pixels, positive correlations were found between N_d and time spent in the free troposphere, mean air-mass trajectory altitude, ocean mixed layer depth, mean zooplankton concentration, and mean boundary layer wind speed. However, most of these correlations occur because there is a split between group 1 and group 2 clouds, one obvious example of this being time spent in the free troposphere.

Differences between groups 1 and 2

The average N_d of group 1 was 78.1 cm^{-3}, while that of group 2 (without island influence) was 47.6 cm^{-3}. The two groups also had statistically significant differences in almost all of their explanatory variables. This leads to the dataset showing statistically significant, if weak, correlations between N_d and all explanatory variables when looking at both groups together. However, separately, the groups exhibit different correlations to these.

Group 1 showed a positive correlation between N_d and time spent in the boundary layer, as well as a negative correlation with mean altitude, which conflicts with the results for the overall group. Wind speed at the cloud pixel also shows a robust negative correlation with N_d for this group, as well as average boundary layer wind speed for days 1-3. Mean sea surface salinity was also a negative influence, however, all group 1 trajectories had a narrow range of 33.8-34.2 PSU, as opposed to the overall dataset spread of 33.5-35.2 PSU, which might make this result less significant. Surprisingly, chlorophyll amount did not show strong consistent correlations with N_d, nor did any other biological variables, aside from a couple that were not consistent for more than two days.

Group 2 showed no correlation between N_d and time spent in the free troposphere or boundary layer, but it did show a consistent negative correlation with mean trajectory altitude, the opposite of the result from the overall data group. Group 2 also showed a consistent positive correlation between N_d and mean boundary layer wind speed, as well as a negative correlation with sea surface salinity. As for biological variables, group 2 showed a negative correlation with chlorophyll concentration, but positive correlations with zooplankton concentration and the mean depth of the euphotic zone.

Predicting N_d from variables

Using a multilinear regression, mean altitude, boundary layer wind speed, chlorophyll concentration, salinity and wind speed at cloud can predict N_d with an accuracy (R^2) of 56%.

Discussion and Implications for Climate Modeling

Trajectories split into two clear groups in this back-trajectory analysis. The main difference between groups 1 and 2 was the altitude at which the air masses spent their final two days. Group 1 spent their final days in the free troposphere and had overall higher N_d, so it is surprising that time spent in the free troposphere was not correlated to N_d in group 2 clouds, despite many group 2 trajectories spending over 48 hours in the free troposphere overall in the 5 days before ending up in a cloud. Group 1 had significantly higher cloud droplet number concentrations than group 2. This suggests that free tropospheric processes are important for the growth of particles to CCN sizes, or that removal mechanisms of marine biogenic aerosols are more abundant in the marine boundary layer.

The relationship, or lack thereof, between mean chlorophyll concentration and N_d is puzzling, as past research has identified a correlation between marine biogenic aerosol and phytoplankton over varying time scales (Ault et al., 2013; Dasarathy et al., 2021; Dror et al., 2018; Gabric et al., 2005, 2002; Lana et al., 2012; McCoy et al., 2015; Meskhidze and Nenes, 2010, 2006; Rinaldi et al., 2010; Sanchez et al., 2021). Chlorophyll concentration at the trajectory location 4 or 5 days before encounter with a cloud does seem to have a positive correlation N_d in both group 1 and 2, but averages over the trajectories do not. This is consistent with DMS emissions, which would not influence N_d without first oxidating to form sulphuric acid and growing particles or nucleating, which can take anywhere between a couple days to a couple weeks (Asher et al., 2011; Becagli et al., 2022; Chen et al., 2018; Fung et al., 2021; Korhonen et al., 2008; Pandis et al., 1994; Stefels et al., 2007; Vallina and Simó, 2007). It is also possible that variations in phytoplankton abundance are less important in the summer, as overall phytoplankton abundance is high, meaning that there is a constant background of CCN and CCN precursors, and the controlling factor of N_d is where these precursors spend their time.

Sea surface salinity and wind speed also seemed to be good predictors of N_d. This may be because sea salt aerosol emissions are controlled by these factors. It is important to note that in this analysis, wind speed correlated negatively with sea surface salinity, so the lack of diverse combinations of values could lead to some inaccurate or overlapping correlations.

The presence of sea salt aerosol can either supplement N_d by acting as CCN, or reduce N_d by taking up water vapour and thus reducing supersaturation, depending on other CCN abundances (Fossum et al., 2020). In the case of group 1, both wind speed and sea surface salinity correlated negatively with N_d, consistent with a scenario of high amounts of non-sea salt CCN. The addition of sea salt aerosol to this scenario will lower N_d by lowering the in-cloud supersaturation, thus leading to a negative correlation between N_d and factors increasing the amount of sea salt aerosol production. Group 2 showed an increase in cloud droplet number concentration with increasing boundary layer wind speeds, with this correlation having a steeper slope for higher chlorophyll values. This relationship is consistent with the wind-speed dependence of sea-spray aerosol, with the increase with chlorophyll exposure explained by the enrichment of sea spray with biogenic aerosol, which is also implemented in the UKESM1 (Mulcahy et al., 2020). Group 2 also showed a decrease in cloud droplet number concentration with increased salinity, which may be due to the aforementioned sea salt reducing supersaturation effect, or the fact that higher wind speeds in the analysis corresponded to lower salinities, and this is simply a coincidence.

Caveats

This analysis sampled clouds on only one day during the southern hemisphere summer, and is therefore limited to the meteorological conditions on the day, as well as high biogenic activity. It is possible that the addition of chlorophyll to a biologically unproductive region will result in an increase in N_d, or that in the winter, continuous time in the free troposphere will have less of an effect on N_d.

The cross-correlation and low variability of some of the trajectory variables also makes it difficult to draw robust conclusions. The next step in this project is to collect data with a higher variability of explanatory variables, so that robust individual relationships between N_d and explanatory variables can be established.

Although this analysis tracked trajectories of individual particles, it is impossible to use these to describe each individual aerosol particle and the reactions they undergo, especially the multiphase chemistry of DMS and its products. This is why it is important to follow up this study with a model study.