The Influence of Aerosols on Cirrus Clouds during an Aircraft Campaign

Introduction & Motivation

Climate change is disrupting societies and harming the delicate ecosystems on Earth (IPCC, 2023). Unfortunately, global-scale action does not seem to be taking place to curb climate change, thus it is necessary that we appropriately evaluate all the potential approaches to mitigate it (Lawrence et al. 2018). One such approach requires the use of an efficient ice nucleating particle (INP) to thin ice clouds, also referred to as cirrus clouds, which are ubiquitous in the atmosphere and uniquely have a net warming effect on Earth due to trapping outgoing longwave radiation (OLR) (Sassen et al., 2008).

Cirrus clouds are composed entirely of ice crystals and form at high altitudes where temperatures can get colder than 235K. Thus, by 'thinning' them with an INP, such as with mineral dust, their optical thickness can decrease allowing OLR to exit the atmosphere and causing a cooling effect on the climate (Mitchell & Finnegan, 2009; Storelvmo et al., 2014). But the success of this approach is reliant on the way that the ice crystals nucleated in the cloud. Ice crystals are formed by either homogeneous or heterogeneous nucleation, the former produces ice crystals that are both numerous and small while the latter creates fewer ice crystals that are larger in size (Storelvmo, 2017). These difference in the ice crystal size distribution controls whether the cirrus cloud has a warming or cooling effect on the climate. Adding an INP to a cirrus cloud formed heterogeneous nucleation would then increase its optical thickness resulting in a warming effect as ice crystals will preferentially form upon the extra INPs rather than growing on the already available ice crystals. Therefore, it is imperative that a method is developed in order to understand the spatial patterns for the two cirrus cloud nucleation formation mechanisms.

Data & Methods

For this project I am using meteorological, cloud, and aerosol measurements from the Atmospheric Tomography Mission (ATom), an airborne field campaign with wide spatial coverage (Wofsy et al., 2021). The flight tracks, separated by each season, for this campaign can be seen in Figure 1. The Cloud Aerosol and Precipitation Spectrometer (CAPS) provides cloud indictor data which is a categorical variable that can be a value from 0 to 4 (0 = cloud free, 1 = aerosol cloud transition regime (ACTR), 2 = liquid cloud, 3 = cloud in the mixed-phase temperature regime, and 4 = cirrus cloud). I plan to use a model to predict this variable with aerosol measurements from the Aerosol Microphysical Properties (AMP) instrument suite which includes the aerosol sizing instruments: 5-chan CPC (N-MASS), ultra-high sensitivity aerosol size spectrometer (UHSAS), and the laser aerosol spectrometer (LAS). These instruments sized dry aerosols encountered during the campaign into the fine (0.0027 um - 0.5 um) and coarse (0.5 um - 4.8 um) size ranges based on the ammonium sulfate optical equivalent diameter.

To begin, I created a feature matrix composed of the time, season, latitude, longitude, altitude, temperature (T), vertical velocity (w), saturation vapor pressure with respect to water, saturation vapor pressure with respect to ice, fine dry aerosol number concentration, coarse dry aerosol number concentration, fine dry aerosol surface area concentration, coarse dry aerosol number concentration, and total aerosol number concentration. I cleaned the data by replacing the fill in value of -99999 with nan and then removing all the rows that contain a nan in any one of the features. After cleaning the data, I was left with 1211447 samples as seen in Figure 1. Next, I also removed any samples that were either cloud free or in the ACTR because I was only interested in the in-cloud data (2 to 4), which left me with 18980 samples (Figure 1). The histograms for each step of cleaning or filtering the dataset from its original number of samples is shown in Figure 2.

Next, I created a feature matrix that only had aerosol measurements in order to predict the in-cloud indicator data. I decided to use a supervised model because I had labeled data as seen in the probability density function (PDF) for each cloud type binned against T (Figure 3). I attempted to use linear SVC, because I was predicting a category and had less than 100K samples, but it did not converge despite increasing the number of maximum iterations to 10K every 10^i starting from i = 1. This demonstrated to me that the variables were too colinear for this type of model, so I opted to use a Support Vector Classification (SVC) with the Gaussian Radial basis function (RBF) instead, which did converge.

Now that the relationship between aerosols and clouds was established, I sought to go one step further and use a model that would use data identified as cirrus cloud by the cloud indicator to establish whether it was formed homogenously or heterogeneously. For this I did not have labeled data, so I needed to turn to a clustering algorithm, an unsupervised method, to find patterns across my feature matrix. Cirrus cloud formation ice nucleation mechanisms are highly dependent on the T, relative humidity, w, aerosol number concentration and aerosol surface area concentration. For homogeneous ice nucleation to take place for cirrus clouds, the T needs to be colder than about 235K and the relative humidity with respect to ice greater than approximately 140%. This is a high energy threshold to be crossed, but as previously mentioned, in the presence of an INP this threshold can be lowered as heterogenous ice nucleation can take place at warmer temperatures and a lower relative humidity with respect to ice. This can be observed in the PDF for cirrus clouds in Figure 3 which is not limited to temperatures colder than 235K. The w is also an important feature as it can uplift water vapor or even ice crystals to colder regions in the upper atmosphere. Specifically, I know that I only have 2 categories (homogeneous and heterogeneous) and that I have less than 10K samples. Thus, I decided to use the K-means model to predict these categories based on these nine features. Before I did the K-means I scaled, or standardized, the feature matrix in order to diminish the sensitivity for different scales across the various features during the clustering process. Then I rescaled the data along with the cluster centroids to plot each feature used in the K-means against T for reference (Figure 4).

Finally, I wanted to predict the locations of these ice nucleation mechanisms, so I attempted to use the labels identified by the K-means clustering algorithm to train a multiple output regression model that predicts latitude and longitude. To identify which label represents which ice nucleation mechanism, I plotted Figure 5 which has two PDFs based on the labels from the K-means clustering against T. I decided that the PDF at warmer temperatures was most likely heterogeneous nucleation while the one at colder temperatures was homogeneous nucleation. Using this information, I set up two feature matrices and target variables for samples identified as homogeneous nucleation and heterogeneous nucleation, respectively. For the feature matrices, I kept the time, month, year, T, w, saturation vapor pressure with respect to water, saturation vapor pressure with respect to ice, fine dry aerosol number concentration, coarse dry aerosol number concentration, fine dry aerosol surface area concentration, coarse dry aerosol number concentration, total aerosol number concentration and the season. The season was encoded from a string into a number that corresponds to each season. Then I used a multiple output regression model with the Random Forest Regressor to predict the latitude and longitude of these two ice nucleation mechanisms.

Results & Limitations

For my first model I used the SVC with a RBF kernel in order to predict cloud types based on a feature matrix with only aerosol measurements. I obtained an accuracy of 74% as seen in Table 1 which shows the classification report for this model. This report provides valuable insights into the model's performance across different classes. Precision represents the accuracy of positive predictions among the instances classified as positive while recall measures the ability of the model to identify all relevant instances of a particular class. The F1-score provides a balanced metric for model evaluation by combining the precision and recall. Looking at the specific class results, the model exhibits high precision of 79% for class 2.0, indicating a low rate of false positives. However, class 3.0 shows lower precision of 69%, suggesting a higher rate of misclassifying instances as positive. Class 4.0 demonstrates a trade-off between precision and recall, resulting in a relatively balanced F1-score 82%. The overall accuracy of 74% indicates the proportion of correctly classified instances across all classes. The macro-average and weighted-average metrics consider either equal weight for each class and account for class imbalances, respectively. Overall, the values for the classification report in Table 1 suggest a reasonably good model performance, especially considering the balanced F1-scores and the weighted average metrics, thus, demonstrating that aerosols can adequately predict the different types of clouds.

I evaluated the K-means clustering by calculated the silhouette score, which is a metric, ranging from -1 to 1, that shows the goodness of a clustering technique. A high value indicates that the object is well matched to its own cluster and poorly matched to neighboring clusters. I obtained a silhouette score of about 0.26. This suggests that the clustering is somewhat reasonable, but there is room for improvement by either using another clustering method or maybe updating the features. The Root Mean Squared Error (RMSE) values for the multiple output regression model were 45.28 and 84.79 for homogeneous and heterogeneous nucleation, respectively. The RMSE is a measure of the average magnitude of the residuals, representing the differences between the predicted and actual values. Lower RMSE values are desirable, as they signify smaller prediction errors and, consequently, higher precision in the model's output. Unfortunately, the RMSE values for this model are high indicating that there are areas for improvement. It is interesting to note that the RMSE is lower for homogeneous nucleation, which actually has more samples (5344) compared to heterogeneous nucleation (3434). Thus, this could be an issue of simply not having enough samples.

There are some limitations to this work. Firstly, cleaning and filtering the original dataset to just in-cloud and cirrus cloud data decreased the number of samples from 1490913 to 18980 and 9154, respectively. As mentioned, I believe it would have been better to have some more samples, especially for the cirrus clouds, in order to train the model. I also believe that for the K-means clustering I did that there was a bit too much collinearity as seen in Figure 4 between T and the supersaturation pressures. Additionally, I encountered some issues with the relative humidity with respect to ice measurements and was not able to use it in the feature matrix. This variable is preferable to saturation vapor pressure with respect to ice because of ease of interpretation, but unfortunately it decreased the number of samples for in-cloud data for summer to none. Finally, the aerosol measurements I used in this project are not from a full-size distribution which paints an inaccurate picture of aerosols in the atmosphere as mineral dust is a coarse aerosol (Kok et al., 2023). This is because the coarse size range upper limit is only 4.8 um and doesn't capture larger sizes of dust which could be better at predicting heterogeneous nucleation. This is not ideal because surface area directly controls ice nucleation as larger INPs can have a higher occurrence of ice activation sites as there is simply more space on its surface.

Future Work & Conclusion

If possible, I plan to refine and extend this work further in the future. I am interested in adding other sources and types of data to get more spatial and temporal coverage. I also would like to narrow down this work and only consider mineral dust as it is an efficient INP that can have implications for climate geoengineering (Cziczo et al., 2013; Lohmann and Gasparini, 2017). As seen in Figure 1, the ATom campaign does not include many samples over Africa, Europe, and Asia, which are important source regions of aerosols. Some deserts in these regions even have deserts that are great at supplying mineral dust into the upper troposphere (Froyd et al., 2022). Unfortunately, there is a lack of mineral dust concentration measurements at cirrus cloud forming altitudes, but it is possible to use ice crystal number concentrations (ICNC) to estimate this value (Burrows et al. 2022). Field campaigns that have captured hydrometeor imaging from the 2D-S Stereo Probe (2DS), which is an optical imaging instrument that obtains stereo cloud particle images and concentrations using linear array shadowing, could provide a promising avenue to obtain this information. For example, a group used hydrometeor images captured by the 2DS instrument during the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES) campaign to discriminate between liquid and ice in the mixed-phase cloud regime by using a random forest algorithm (Atlas et al., 2021). I would be interested in using a similar method to discriminate between ice crystals that were formed heterogeneously with the assistance of an INP and those that were formed by homogeneous freezing. However, it is important to note though that this may cause some overestimation issues in the derived mineral dust concentration due to secondary ice production processes being difficult to account for.

There are a few machine learning techniques I would also like to try for this work. The silhouette score was low for the K-mean clustering, so I think it would be beneficial to further consider what features I am using for that particular model. Maybe it would be possible to do a principal component analysis in order to figure out what features are redundant and reduce the feature matrix dimensionality to improve on the clustering. I also would be interested in performing a hyperparameter tuning using Grid Search for the multiple output regression model. This would improve the model by narrowing down the parameters that can reduce the RMSE.

Ultimately, this research could eventually inform cirrus cloud geoengineering by indicating what regions on Earth would be best suited for introducing mineral dust to cirrus clouds in order to produce a cooling effect. Given the current state of climate change it is necessary to conduct this research now especially given that geoengineering as a whole has been largely ignored in practice due to its uncertain consequences. But, it is still important that we properly assess whether this method could work along with the risks associated with it, so that humanity can make a well-informed decision as climate change continues to impact life on Earth.

Table 1 | *A classification report for the SVC with RBF analysis.*

	Precision	Recall	F1-score	Support
2.0	79%	79%	79%	1047
3.0	69%	20%	31%	796
4.0	73%	95%	82%	1820
Accuracy			74%	3663
Macro Average	74%	65%	64%	3663
Weighted Average	74%	74%	70%	3663

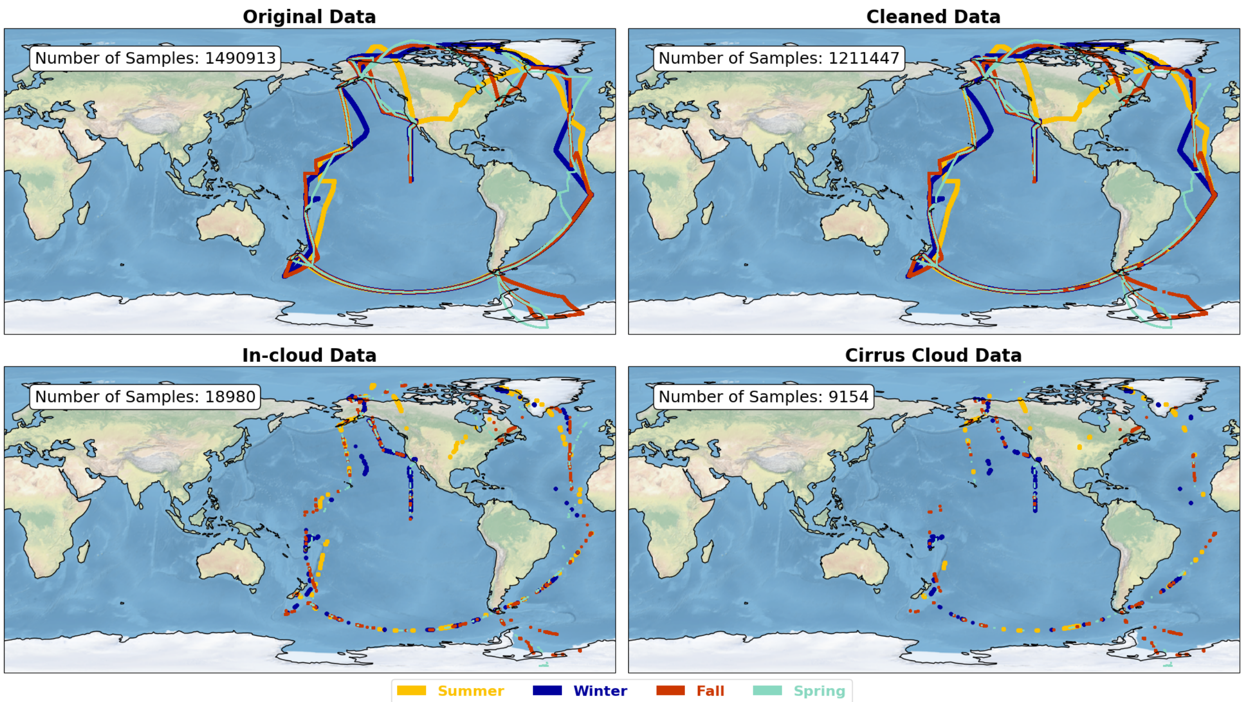

Figure 1 | The flight tracks for the ATom campaign for each season separated by original data, cleaned data, in-cloud data, and cirrus cloud data.

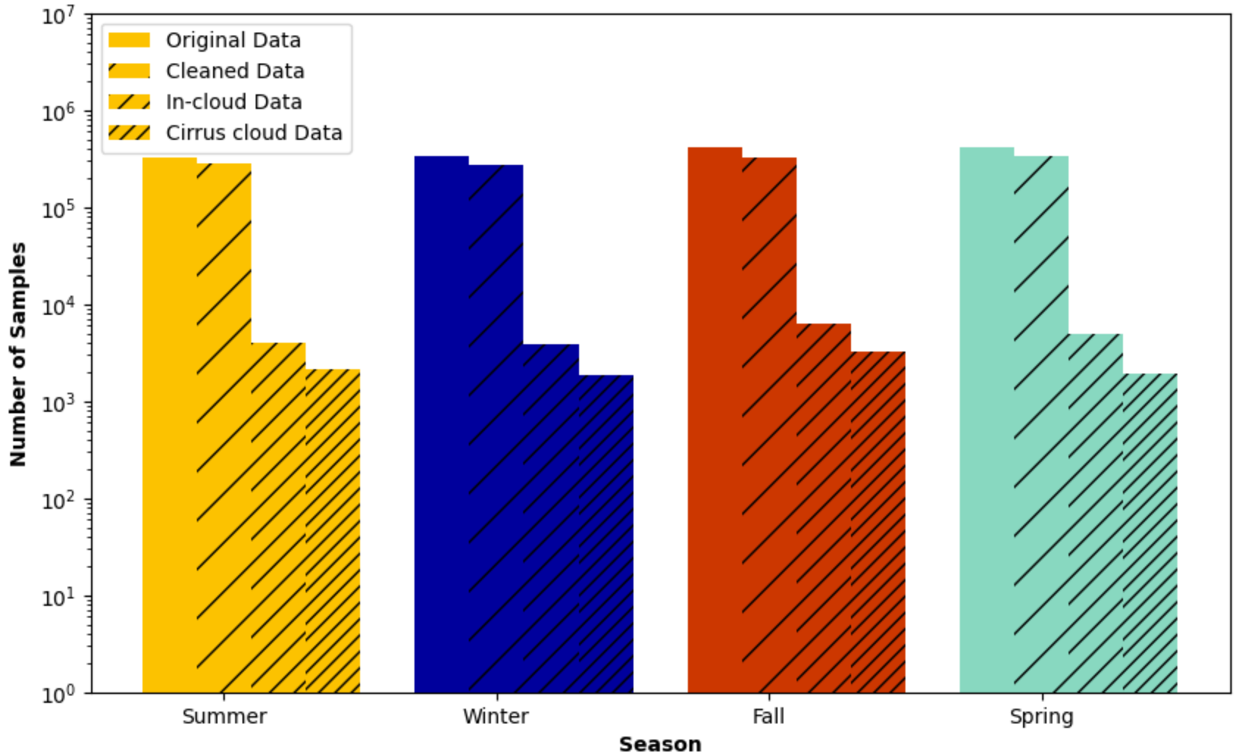

Figure 2 | Histograms that show the number of samples for each season in the ATom campaign further separated into the original data, the cleaned data, in-cloud data, and cirrus cloud data.

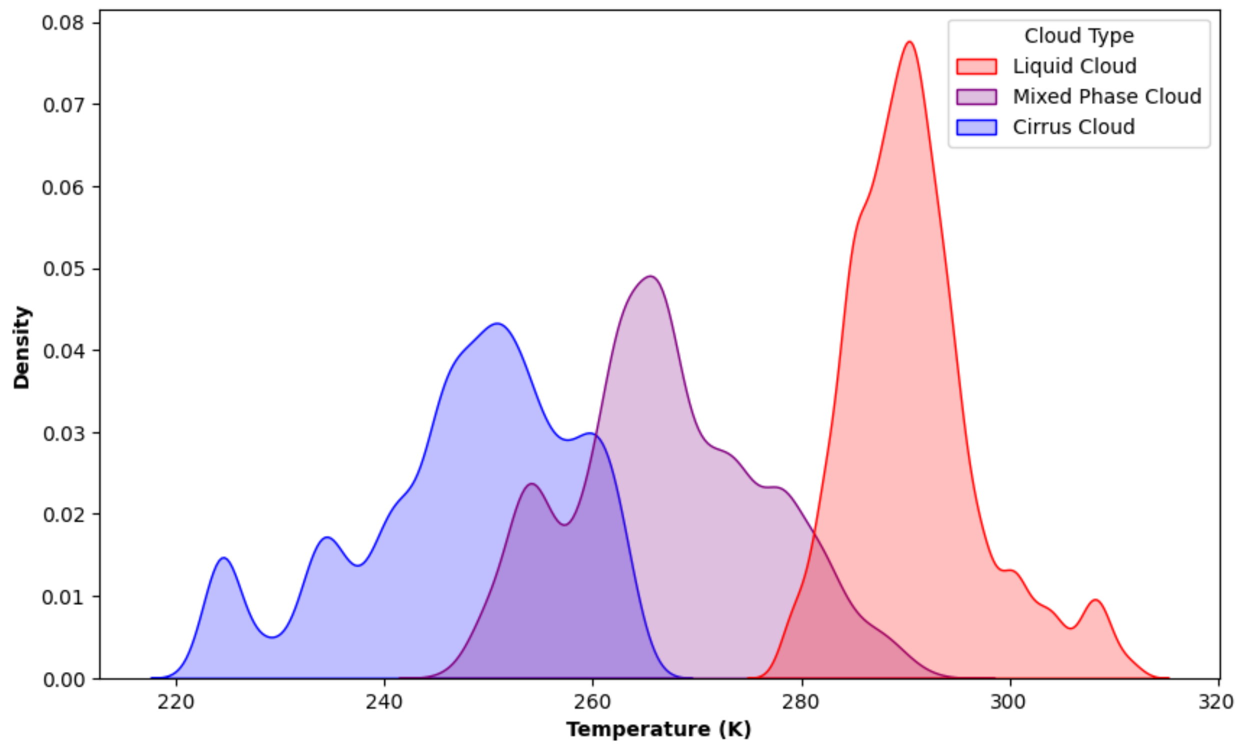

Figure 3 | Probability distribution functions for the three types of cloud in the ATom dataset

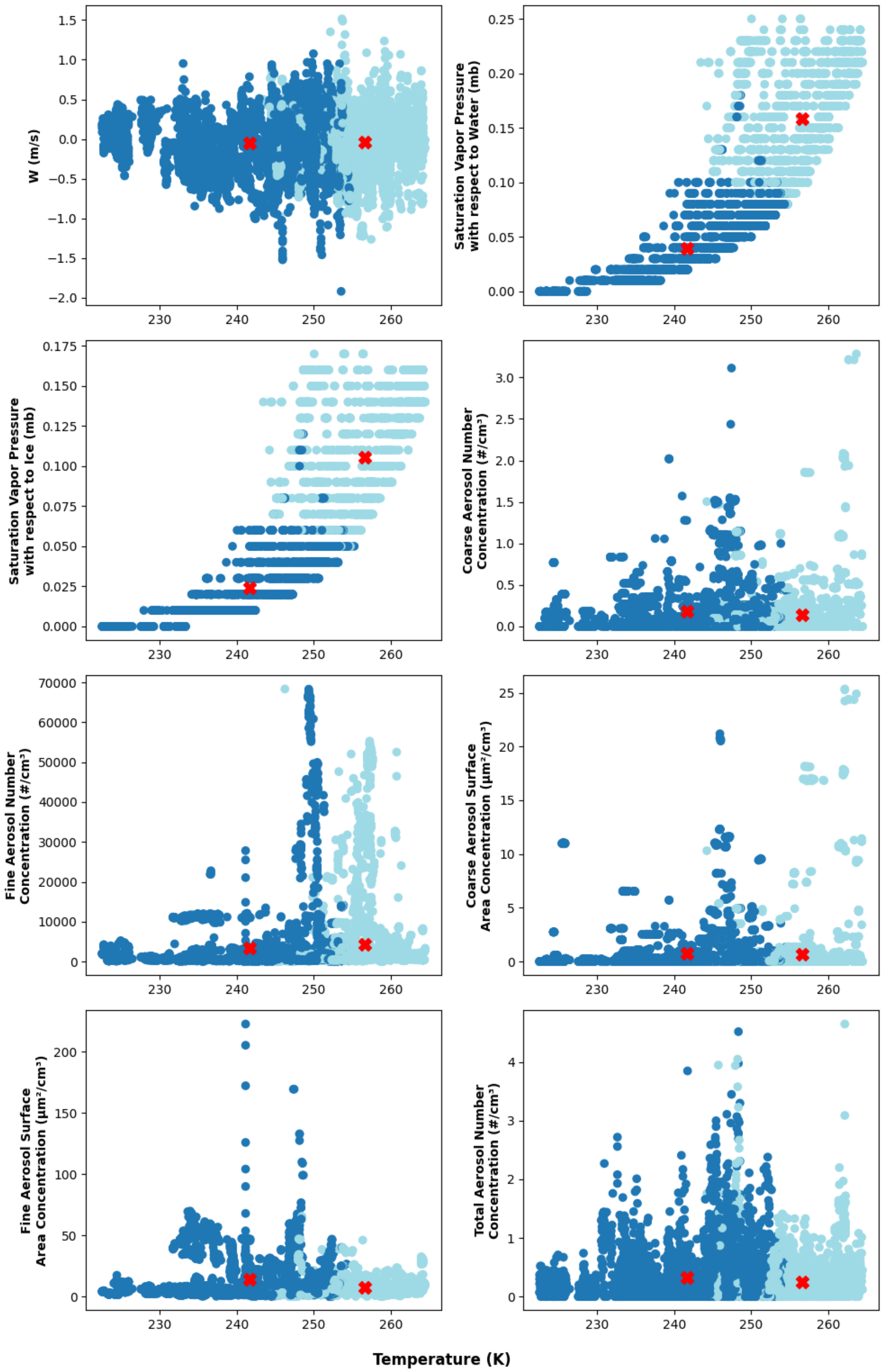

Figure 4 | Scatter plots for the features used for the K-means clustering plotted against T. Red Xs signify the cluster's centroid.

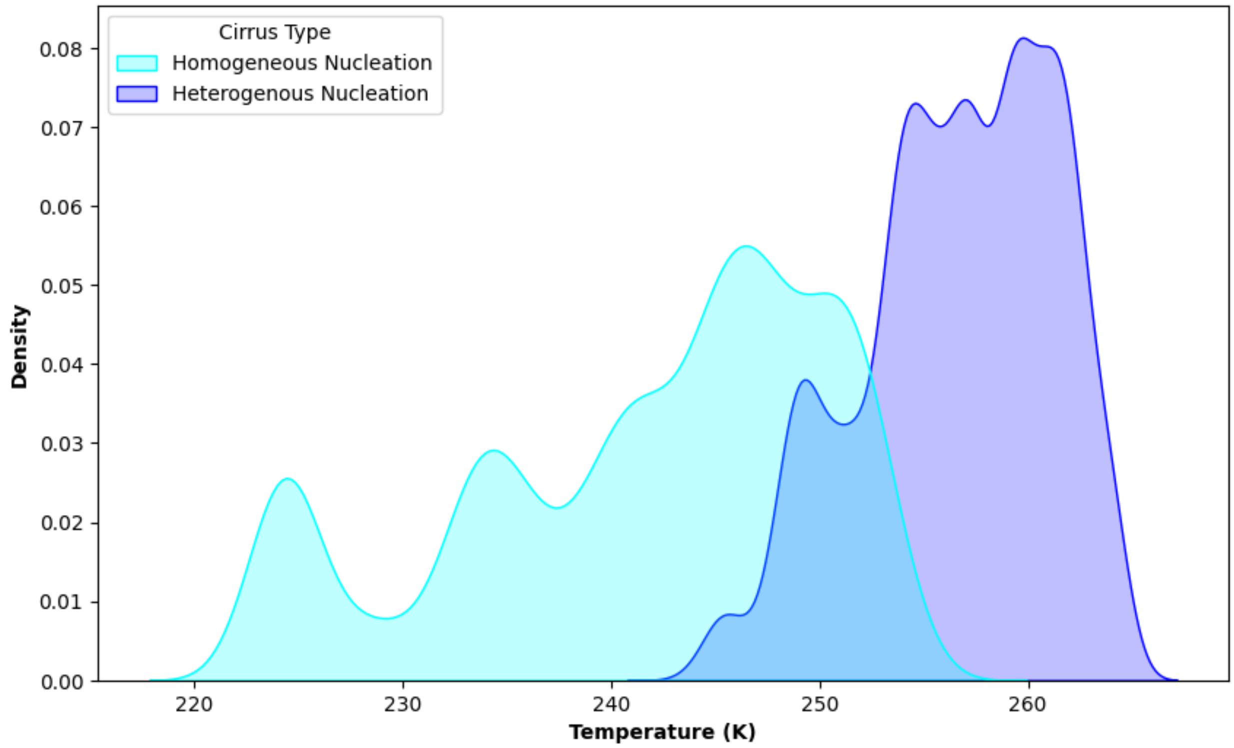

Figure 5 | Probability distribution functions for the two ice nucleation formation mechanisms for cirrus clouds as identified using K-means clustering.

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