

A06: Analysis of GRAPE data

The effects of anthropogenic aerosols on cloud microphysical properties.

Candidate Number: 32835

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Abstract: Space-borne retrievals of aerosol optical depth, cloud optical depth and cloud droplet effective radius from the second Along-Track Scanning Radiometer instrument (ATSR-2) are used to explore the effect of aerosol on the microphysical properties of clouds. Two regions are studied: one off the African coast and the other off the Japanese coast. In the former region the effects of an annual biomass-burning event are studied, in the latter the effects of industrial and urban aerosols are investigated. A third ‘aerosol-free’ region is considered as a control. In both of the study regions an increase in aerosol loading is observed to cause a decrease in the mean cloud droplet size, in agreement with the Twomey ‘first indirect effect’ hypothesis. A parameterisation of the Twomey effect is applied in each region and the results show a good agreement with cloud process models. The cloud optical depth and aerosol optical depth are also found to be related, particularly in the region off the African coast.

1 Introduction

Over the past few years, extensive studies have been carried out into the interaction of clouds and aerosols using computer models, in situ measurements and remote sensing observations[1][2][3]. It is observed that small (sub-micron scale) aerosol particles have a profound effect on the Earth’s climate. Such particles modify atmospheric scattering and absorption processes, in turn altering the radiative balance of the atmosphere. This effect is known as the ‘first direct radiative forcing’[4] and in general tends to reduce the mean surface temperature. Dark-coloured aerosol particles absorb sunlight and thus warm the atmosphere relative to the surface. It is predicted that the net effect of this warming is to reduce the upward movement of moisture and in turn reduce the cloud cover[5]. This ‘semi-direct’ effect reduces the Earth’s albedo, and thus increases the mean surface temperature.

Atmospheric aerosols can also act as cloud condensation nuclei (CCN) and thereby increase the number of droplets forming within clouds. In general, this tends to bring about a decrease in the mean droplet size[6], which causes an increase in the short-wave reflectance¹. The net effect of aerosol loading on albedo of clouds is known as the ‘first in-

direct effect’, or the ‘Twomey effect’[7]. Through secondary ‘indirect effects’, aerosols can suppress precipitation and increase cloud lifetimes[8][9][10]. These processes tend to cool the Earth, reducing the impact of global warming.

Industrial and agricultural human activity is responsible for a significant amount of the aerosol in the atmosphere[11]. Although aerosols from such activity are typically hydrophobic, recent research indicates that as they age in the atmosphere they can become sufficiently hydrophilic to act as CCN and thus give rise to indirect effects[12]. It is therefore clear that anthropogenic aerosols can have a profound impact on the radiative properties of clouds, through the action of both the ‘direct’ and ‘indirect’ effects.

Clouds play a critical part in the Earth’s climate system, because of both their radiative properties and their role in the hydrological cycle. Accurately quantifying cloud effects is therefore a vital part of producing a model of the Earth’s climate[13]. In 2001, estimates of cloud indirect effects due to anthropogenic aerosols were identified by the Intergovernmental Panel on Climate Change (IPCC) as one of the greatest uncertainties in predictions of climate change[14]. This is still the case today[15]: the first working group of the 2007 IPCC[16] have recently concluded that estimates of the ‘cloud albedo effect’ remain a large uncertainty in climate modelling

¹ The presence of aerosol can cause a decrease in the mean droplet radius for clouds of very high optical depth.

studies, describing the effect as an area of which scientists have a ‘low level of scientific understanding’. It is very important that further research into indirect effects is carried out in order to reduce these uncertainties and to improve our understanding of the climate and the possible effects of anthropogenic aerosols[17][2].

The aim of this report is to quantify the ‘first indirect effect’ on the size of cloud droplets due to the regional loading of aerosols from biomass burning and industrial/urban pollution.

2 The instrument

The second Along Track Scanning Radiometer (ATSR-2)² is ideally suited for an analysis of the effect of aerosol on cloud properties. ATSR-2 images the Earth in seven wavelength bands: four visible and near-infrared channels and three thermal infrared channels[18]. The instrument provides a forward view (at a zenith angle of $\sim 56^\circ$) and a nadir view at each point. The swath width over the land is nominally 512km, but over the ocean it is reduced to 256km, due to bandwidth limitations of the ERS-2 satellite on which it operates. ATSR-2 was launched on the 21st of April 1995, but went out of service between December 1995 and May 1996 due to a problem with the scanning mirror. In 2001 a fault developed with the alignment mechanism on the ERS-2 satellite, and so at present no data is available from after that date³. The current ‘level 1’ data set therefore comprises five years of operation.

3 The data

The satellite data is processed at the Rutherford Appleton Laboratory using the GRAPE algorithm⁴[19], which uses an optimal estimation technique⁵ to evaluate cloud or aerosol properties. First of all, a ‘cloud flagging program’ is used to decide whether a particular set of measurements corresponds to the instrument viewing cloud or aerosol.

Then, for each measurement, a forward model is used to predict the radiances that would be expected in each wavelength channel, using a number of cloud and aerosol parameters⁶. Different forward models are used for retrievals of cloud and aerosol. A cost function is calculated based on the weighted mean-square difference between the measured radiance values and those predicted by the forward model as well as a priori parameter information. The procedure is iterated until this cost function is minimised. This way, a set of either aerosol or cloud parameters is built up for each pixel. Further parameters, such as cloud top height are calculated using temperature profiles calculated at the European Centre for Medium-Range Weather Forecasting (ECMWF)⁷. Using the optimal estimation technique, it is possible to calculate rigorous error estimates on each of the derived parameters, which are important when filtering and analysing the data. A flow diagram of the retrieval process is given in figure 1.

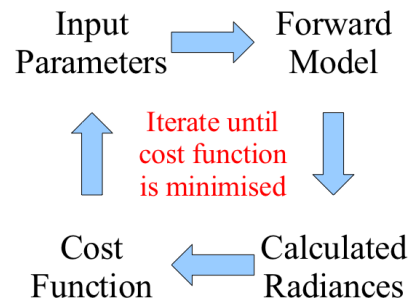


Fig. 1: The GRAPE retrieval process, after cloud flagging has been applied.

The parameters in the retrieval include aerosol optical depth, cloud fraction, cloud effective radius, cloud top height and cloud optical depth. The retrieved data is stored in two different formats: ‘level 2’ data files store information for each individual orbit on a pixel-by-pixel basis, whereas ‘level 3’ data files contain monthly-averaged data stored in 2.5° latitude/longitude boxes, with filtering applied to discount contributions from any erroneous pixels.

² For more information on ATSR-2, visit the project website: <http://www.atsr.rl.ac.uk/index.shtml>.

³ Recent work to map the data from 2001 onwards is showing significant signs of success; features such as land ocean boundaries are used to identify the origin of particular measurements and thus calculate the alignment of the satellite.

⁴ The GRAPE algorithm was developed at Oxford University and the Rutherford Appleton Laboratory. For more information on the GRAPE processing, visit <http://www.atm.ox.ac.uk/project/grape/index.html>.

⁵ For a rigorous introduction to inverse methods and estimation theory, refer to the book by Rodgers[20].

⁶ The forward model incorporates estimates of the surface reflectance of the Earth. Over the ocean these estimates are calculated using Cox and Munk statistics[21], whereas over the land, a bi-directional reflectance distribution function, derived from MODIS retrievals, is used. For more information on the Moderate Resolution Imaging Spectroradiometer (MODIS) visit <http://www.modis.gsfc.nasa.gov>.

⁷ At the ECMWF, atmospheric data from many sources is used in combination with a general circulation model (GCM) to calculate global atmospheric temperature profiles. For further information visit <http://www.ecmwf.int>.

The GRAPE processing is not yet complete⁸; continuous level 2 and 3 data is currently available from mid-1996 to mid-1998.

4 Regions of study

Initially, a plotting program was applied to the level 3 data to produce maps of monthly mean aerosol optical depth (AOD) and cloud effective radius (CER). These maps were inspected by eye in order to identify potential regions for further analysis. For each region, a 10° latitude/longitude box was defined, corresponding to an area of $\sim 1100 \times 1100 \text{ km}^2$.

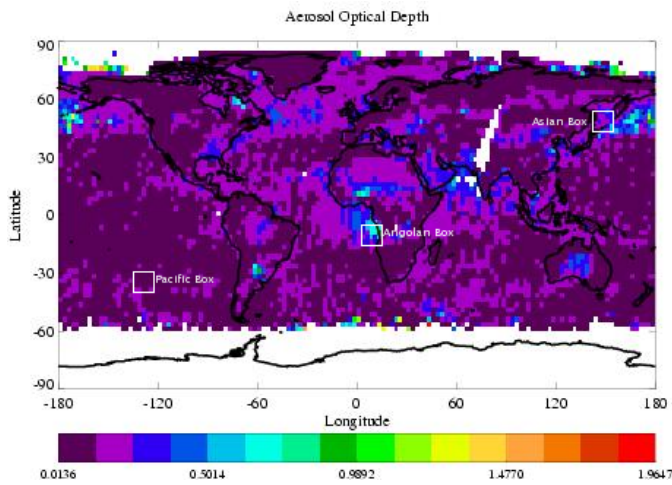


Fig. 2: Mean aerosol optical depth during August 1996. Note that areas of white space correspond to regions in which an accurate retrieval was not possible [usually due to a low sun elevation]. The locations of the case-study regions are also given.

4.1 Angolan coastal region

Figure 2 shows the global monthly-averaged AOD for August 1996. A region of high mean AOD can clearly be observed off the coast of Angola. This region of high aerosol loading can be seen in the monthly-averaged AOD maps for July, August and September each year, but is not visible during the other months. The appearance of this aerosol ‘plume’ corresponds to the crop-burning season in Angola, Namibia and Zaire which generally begins in late July or early August and comes to an end in October[4]. Investigation of the region just off the Angolan coast can thus provide an opportunity to analyse the effects of aerosol from biomass-burning on cloud. The details of the African region chosen are given in the table below:

Angolan Region:

5°	<	Longitude	<	15°
−18°	<	Latitude	<	−8°

4.2 Asian region

In figure 2 it is also possible to identify a region of high aerosol loading to the north-east of Japan and Northern China. The monthly-averaged maps of AOD show that the aerosol loading in this region can reach very high levels. A ‘plume’ of very high AOD appears at around May/June each year. The origin of the aerosol loading in this region is likely to be the out-flow of industrial and urban aerosols from northern China and Japan, since the prevailing (geostrophic) wind in that region of the globe is south-westerly. The fluctuating levels of mean AOD presumably correspond to fluctuations in the winds off the Japanese coast. This area provides an opportunity to analyse the effects of aerosols due to urban and industrial pollution. The details of the Asian study region are given in the table below:

Asian Region:

150°	<	Longitude	<	160°
45°	<	Latitude	<	55°

4.3 Pacific region

It is clear in figure 2 that in the Southern Pacific the aerosol optical depth is very low throughout the year. In this region it is possible to investigate clouds that have developed in an atmosphere characterized only by maritime aerosols, far from any anthropogenic aerosol sources. The details of the Pacific study region are given in the table below:

Pacific Region:

−130°	<	Longitude	<	−120°
−30°	<	Latitude	<	−40°

5 Experimental process

5.1 Data Filtering

The ‘level 2’ data comprises the values obtained for each of the retrieved parameters from all of the measurements taken by the instrument in a particular orbit. Individual retrievals can fail to converge and thus produce erroneous parameter values if, for instance, the cloud flagging algorithm does not correctly identify the contents of a pixel. Parameters for which the retrieval has failed to converge are set to an ‘error value’ such as −999. Sometimes, retrievals produce erroneous values due to a very poor fit between the measured radiances and those

⁸ At the time of research; March/April 2007.

predicted by the forward model. Such retrievals will have large values of the cost function, and will only converge after a large number of iterations. anomalous parameter values may be predicted due to weaknesses in the forward model. Highly anomalous parameter values can be filtered out of the dataset by considering prior knowledge, or by considering the rigorous errors calculated on each of the parameter values during the retrieval process.

Anomalous retrievals are filtered from the dataset using simple constraints. For the aerosol retrievals the filtering is as follows:

Parameter	Constraint
Julian date (JDT)	> 0
No. iterations (ITR)	> 0
Cost (CST)	< 10

For the cloud retrievals, more extensive filtering is used:

Parameter	Constraint
Julian date (JDT)	> 0
No. iterations (ITR)	> 0
Cost (CST)	< 30
Cloud top pressure (CTP)	< 1000
Cloud optical depth (COD)	> 2
Error on COD	< 80
Error on CER	< 10
Error on CTP	< 400

5.2 Data extraction

The data extraction program⁹ loads in turn any level 2 data files that correspond to orbits which overlap the specified region. Only the pixels which lie within the specified latitude and longitude limits are considered. Any erroneous parameter retrievals are filtered out using the constraints outlined above, before mean values are calculated for each of the parameters. The parameters investigated are: AOD, COD, CER and cloud top height, CTH. Thus, for each orbit, a value of the regional mean AOD, mean CER, mean COD and mean CTH is stored. The number of measurements contributing to each mean value and the standard deviation of each of the mean values are also stored. This ‘level 4’ data is used as the basis for the analysis in the present work.

5.3 Data processing

It is found that anthropogenic aerosols emitted in the atmosphere tend to lie in the altitude range $\sim 1.8 - 3.7\text{km}$ [22]. Some research also indicates

that further aerosol layers can be found at higher elevations in the atmosphere[23][24]. We would thus like to analyse cloud and aerosol data from specific height ranges within the atmosphere.

Initially, the analysis program¹⁰ considers the extracted ‘level 4’ data from any orbits that contribute more than 500 measurements of each parameter. Any orbits that produce zero mean values for any of the parameters are ignored. The data from each orbit is initially filtered into 1km bins depending on its mean CTH. Then, in each height range, an n-day rolling average through the mean AOD, mean COD and mean CER data can be calculated, ignoring any points which lie more than two standard deviations away from each n-day mean. This process allows any anomalous values, or unexpected fluctuations due to short term weather to be filtered out of the data. A standard deviation is calculated for each n-day average, considering the standard deviations of the constituent averages, using the formula:

$$1/\sigma_{tot}^2 = \sum_i (1/\sigma_i^2)$$

The standard errors are not calculated, since they are, in general, vanishingly small, due to the huge number of measurements taken by the instrument during each orbit.

6 Results

6.1 African region

The results obtained in the African region are summarised in figure 3. The first plot (A) shows the time series of the mean AOD and the mean CER in the region, for the duration of the dataset. Each point in the plot corresponds to data from one orbit, averaged over the entire region. Over the ocean, the satellite swath width is approximately one quarter of the width of the region considered, so each orbit will contribute data from slightly different parts of the ‘box’.

The most striking feature of plot A is the sudden increase observed in the mean AOD at around August each year. These ‘spikes’ in the mean AOD occur when there is a large flux of soot-based aerosol into the region, due to crop-burning activities in Angola, Namibia and Zaire. In August, when the region is heavily loaded with soot, the mean AOD rises as high as ~ 1.2 , compared to a mean value of ~ 0.2 during the remainder of the year. The mean CER does not exhibit such clear variation, but there are nevertheless interesting features in the

⁹ The analysis programs were written using IDL. For an outline of the program code, see appendix I.

¹⁰ For an outline of the IDL program code, see appendix II.

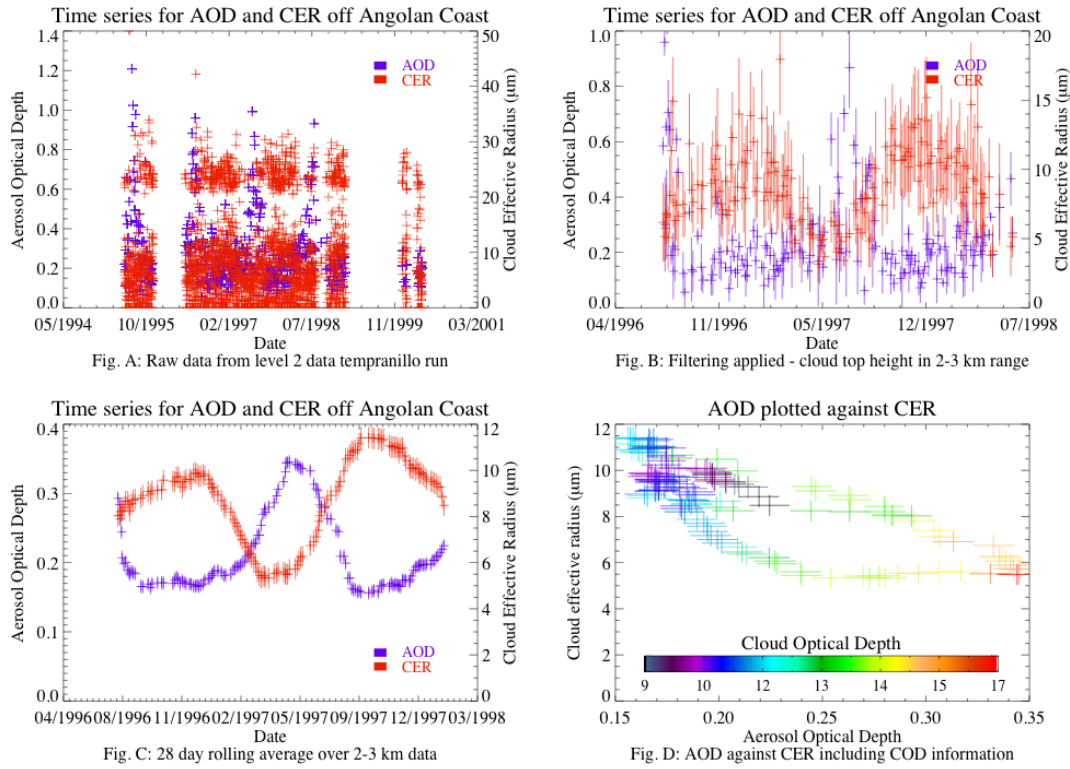


Fig. 3: Results of the analysis off the Angolan coast

data. The mean CER values appear to fall into two 'bands': one band with droplet sizes between $0\mu\text{m}$ and $18\mu\text{m}$ and a second band with droplet sizes in the range $20\mu\text{m}$ to $30\mu\text{m}$. The first band, of higher effective radius, is likely to correspond to ice clouds at higher altitudes ($> 4\text{km}$) and the second band to water clouds at lower altitudes. During the periods of high aerosol loading the number of ice clouds is observed to decrease. There also appears to be a decrease in the mean CER of the lower altitude clouds when the mean AOD increases.

The second plot (B) is of a similar format. Only orbits containing cloud at a mean height of between 2 km and 3 km are considered. This height range, as expected, seems to correspond to the most clear aerosol loading. Clouds in this height range are generally found to consist of water droplets, rather than ice crystals. The region of continuous data from mid-1996 to mid-1998 is plotted. Once again, the annual aerosol loading plume is clearly visible, with the mean AOD increasing as high as 1.0. In this plot however, the effect of this loading on the mean CER is significantly more clear. The mean CER ranges between $\sim 15\mu\text{m}$ when the mean AOD is minimum, to below $5\mu\text{m}$ at the peak of the mean AOD. The error bars on plot B correspond to the standard deviations on the averages taken to produce each data point, thus giving an indication of

the range of values encountered in the region. The spatial variation of both the AOD and CER values is observed to be fairly constant: ~ 0.2 for AOD and $\sim 5\mu\text{m}$ for CER. The points with unusually small error bars are not necessarily more accurate, since they may correspond to a mean taken over a relatively small number of individual retrievals. The points with unusually large error bars are likely to represent the effects of weather, or perhaps contain erroneous retrievals.

Plot C shows the result of taking a 28-day rolling average¹¹ through the mean AOD and CER data from plot B, in the 2-3km height region. The timescale for this average is chosen such that it is smaller than that of the biomass burning period, but is long enough to smooth out any fluctuations in the parameters due to weather (the effects of weather are likely to cause variations on a timescale of a few days). The result is a very clear relationship between the AOD and the CER. The mean CER rises and falls in tandem with changes in the aerosol loading of the region, increasing as the aerosol loading decreases. This is very clear evidence that indirect effects resulting from atmospheric smoke pollution can cause profound changes in cloud properties. Again, the error bars show the standard devi-

¹¹ The averaging process is described in the 'data processing' section of the report.

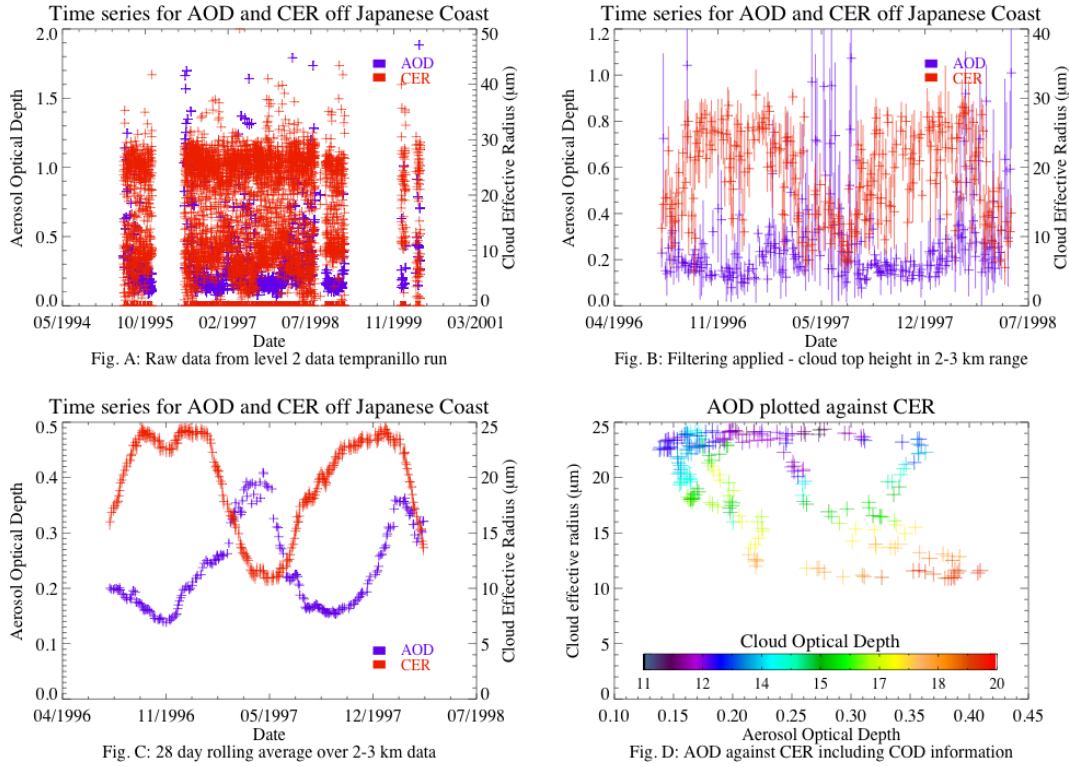


Fig. 4: Results of the analysis off the Japanese coast

ations on the averages taken in producing each data point, giving an indication of the spatial variation of the mean AOD and mean CER values over the 28-day period. The variations in the CER caused by 'weather' are much smaller than the effects of the annual plume of biomass-burning aerosol.

The final plot is a scatter plot of 28-day mean AOD against 28-day mean CER. The points are plotted in different colours according to the corresponding 28-day mean COD, as indicated by the colour bar. The mean AOD ranges between ~ 0.15 and ~ 0.40 and the mean CER ranges between $\sim 4\mu\text{m}$ and $\sim 12\mu\text{m}$. In general, the mean CER is observed to decrease with an increase in mean AOD, as would be expected for an indirect effect. The exact functional form of this decrease is not clear, but the plot indicates a bimodal relationship. The upper branch of the plot appears to behave fairly linearly. The lower branch possibly indicates a saturation of the Twomey effect in this region when the mean CER drops to ~ 5 . In general, the mean COD appears to increase with increasing AOD and thus with decreasing CER, possibly due to water vapour emitted in tandem with the aerosol. Error bars are plotted for both mean AOD and mean CER; the errors plotted are exactly the same as those plotted in the third plot.

6.2 Asian region

The results obtained in the Asian region are summarised in figure 4. The plots A - D are completely analogous to those in figure 3, giving the results in the Angolan region. In plot A, it is possible to discern a sharp increase in the mean AOD each June. This is very likely to correspond to a seasonal change in the weather, increasing the flux of pollution from northern China and Japan into the region. The aerosol optical depth rises even higher here than in the African region, to values in excess of 1.5. Once again, the mean AOD for the majority of the year seems to fall in the range ~ 0.1 to ~ 0.2 . The mean CER shows very little evidence of any reaction to the aerosol loading, although the number of clouds with effective radii in the range $1-5\mu\text{m}$ seems to fall to zero when the mean AOD reaches a maximum. The mean CER values again seem to fall into two 'bands', although the effect is less clear than in the African region.

The second plot shows a clear relationship between the mean AOD and the mean CER for clouds in the 2-3km range. The range and size of the mean AOD values is very similar to that observed in plot B for the African region. The mean CER values in this plot, on the other hand, are much greater than those observed in the African region, perhaps due to some variation of the mean droplet size with

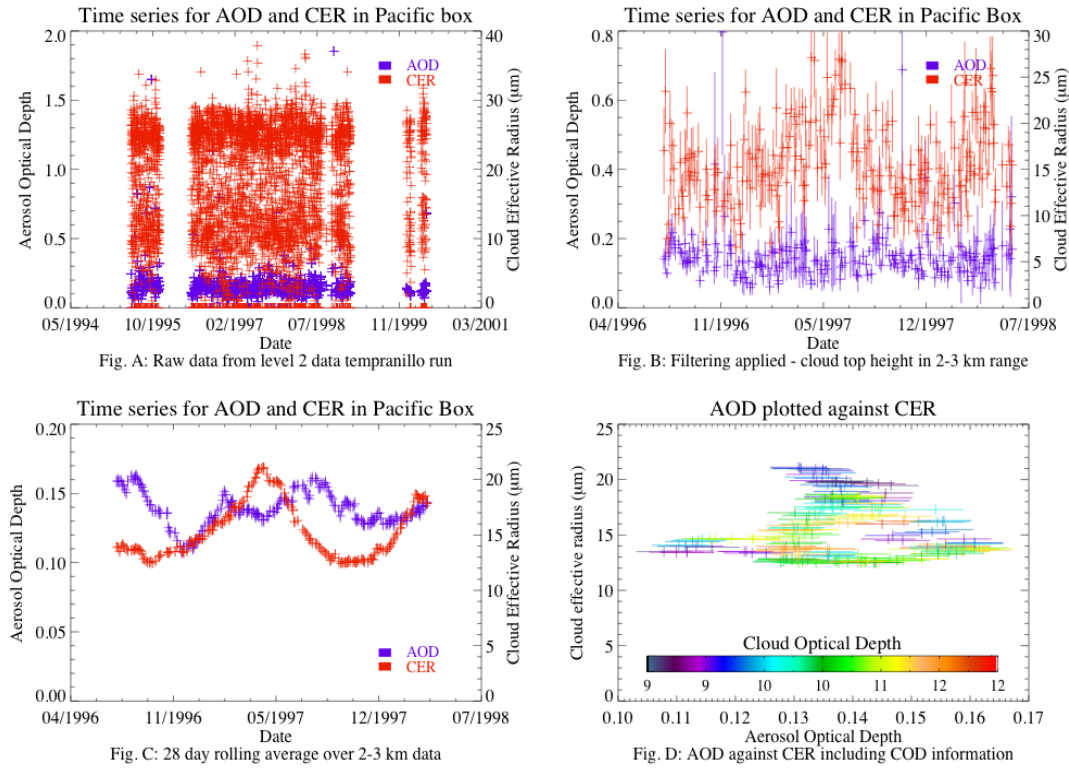


Fig. 5: Results of the analysis program in the Pacific box

latitude. The mean CER has a similar range to that in the African region, decreasing from a value of $\sim 25\mu\text{m}$ when the aerosol loading is at its minimum to $\sim 12\mu\text{m}$ at the peak of the aerosol loading. The ‘error bars’ on the mean AOD, and thus the range of values contributing to the spatial average, appear to increase in size when the aerosol loading is increased. The range of CER values contributing to the regional mean CER appears to be fairly constant and is smaller than the variation of the mean CER over the data period.

Plot C again gives the results of a 28-day rolling mean taken through the data in plot B. As in the African region, this timescale is smaller than that of the period of raised AOD, but long enough to smooth out daily/weekly fluctuations in the dataset. The plot yields similar results to the African region. An increase in the mean AOD clearly results in a decrease in the mean CER and vice versa, in agreement with the Twomey hypothesis. In this plot however the peaks and troughs do not appear to align exactly, but instead the ‘response’ of the mean CER to an increase in the mean AOD seems to trail the aerosol loading by approximately half a month. It is not clear what might cause this trailing effect. The error bars on this plot are very small due to the large number of points, each with a relatively small standard deviation, that contribute

to the mean.

The final plot in figure 4 is not clear. The general trend of increasing mean AOD corresponding to decreasing mean CER is visible, however the plot appears to have three branches, possibly due to the ‘lag’ of the response of the mean CER to the mean AOD. There is a clear relation between the mean COD and the mean CER, with higher effective radii corresponding to lower cloud optical thicknesses. In this region the Twomey effect appears to saturate at a mean CER of ~ 12 although it is difficult to draw a firm conclusion.

6.3 Pacific Region

Figure 5 summarises the analysis performed in the Pacific box. The plots A-D are produced in exactly the same way as those in figures 3 and 4 for the Angolan and Asian regions.

In plot A it is not possible to discern any temporal patterns in the mean AOD or mean CER values. The mean CER values again seem to be broadly distributed in two main bands, but the difference is much less clear than in the African region.

In plot B it can be seen that the aerosol loading in the 2-3km range remains fairly constant at approximately ~ 1.5 . Despite this, there is a clear variation of the mean CER, which seems to peak in May 1997. This indicates that the CER is a not only function

of AOD, but perhaps also has seasonal and spatial dependences. It is interesting to note that at this time of year in the other regions the mean CER is in general low.

In the third plot the 28-mean AOD and CER are both observed to fluctuate by a factor of ~ 0.7 over a timescale of approximately one year. The fluctuations in the mean AOD are a more than an order of magnitude smaller than in either of the other regions; there is no explicit aerosol forcing. The mean CER has a large peak at around May 1997 that does not correspond to a period of significant change in the mean AOD.

Plot D shows no clear trend between mean AOD and mean CER. The prominent behaviour is a straight line of zero gradient, corresponding to fluctuations in the CER with the aerosol loading remaining fairly constant. Neither the mean AOD or the mean CER seem to be related to the mean COD. The mean COD in the Pacific region has a much smaller range than in the African or Asian regions, of $\sim 9 - 12$.

6.4 Analysis

We wish to compare the relationship observed between the mean AOD and the mean CER in the African and Asian regions to the theory proposed by Twomey[7]. According to Twomey's hypothesis, the number of droplets forming within a cloud, N_d , is related to the number of aerosol particles which may act as cloud condensation nuclei (CCN), N_a , through the simple relation:

$$N_d = (N_a)^\alpha \quad (1)$$

According to measurements and modelling studies of clouds[6], we expect $\alpha = 0.7$. We assume that the total volume of water is conserved, giving the relationship:

$$N_d \times \frac{4\pi}{3} r^3 = \text{constant} \quad (2)$$

The total number of aerosol particles in the atmosphere can be assumed to be proportional to the mean AOD. However, we have only considered CER data from orbits which contain cloud at a mean height of 2-3km. In order to quantify the Twomey effect, we wish to calculate the number of anthropogenic aerosol particles that may act as CCN in this height range. First of all, the 'background aerosol', not resulting from human activity, is subtracted from the data, in order to calculate a value for the total amount of anthropogenic aerosol. We

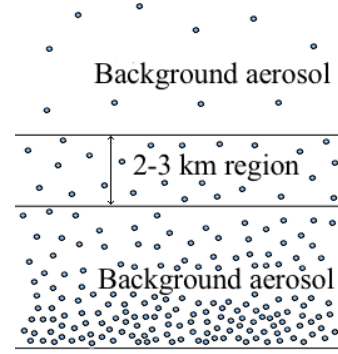


Fig. 6: A crude representation of the 'background' distribution of aerosol particles in the study regions when the aerosol loading is *not* present.

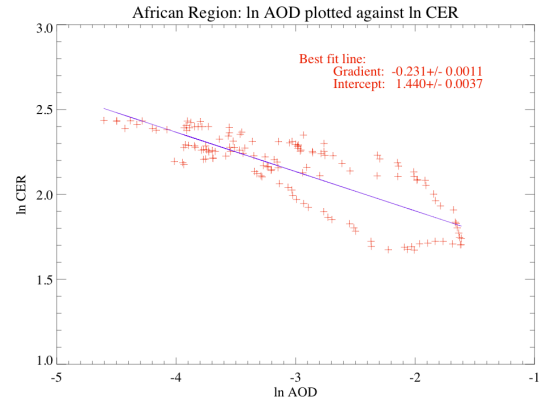


Fig. 7: A log-log plot of mean AOD and mean CER in the African region. A chi-squared linear fit is used to estimate the mean gradient.

assume that the 'background aerosol', which is illustrated in figure 6, corresponds to the minimum value of the 28-day mean AOD observed in the region, over the complete dataset. The number of anthropogenic aerosol particles which may act as CCN in the 2 – 3km range, N_a , is then assumed to be proportional to the total amount of anthropogenic aerosol. We thus have:

$$N_a \propto [\text{total aerosol}] - [\text{background aerosol}] \quad (3)$$

$$\therefore N_a \propto [\text{total AOD}] - [\text{background AOD}] \quad (4)$$

Combining equations 1, 2 and 4 we have:

$$[\text{total AOD} - \text{background AOD}]^\alpha \propto [\text{CER}]^{-3} \quad (5)$$

So, we have:

$$\begin{aligned} \frac{\alpha}{3} \log [\text{total AOD} - \text{background AOD}] \\ = \log [\text{CER}] + \text{constant} \end{aligned} \quad (6)$$

We would thus expect a plot of $\ln(\text{mean AOD})$ against $\ln(\text{mean CER})$ to yield a linear relationship

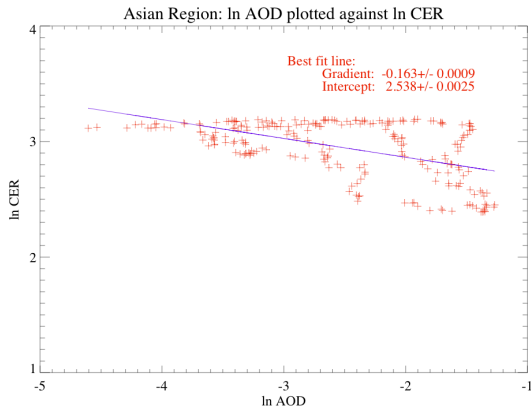


Fig. 8: A log-log plot of mean AOD and mean CER in the Asian region. Again, a chi-squared linear fit is used to estimate the mean gradient.

with a gradient of $\frac{\alpha}{0.7} = -0.23$. Figures 7 and 8 are the relevant plots of $\ln(28\text{-day mean AOD})$ against $\ln(28\text{-day mean CER})$ for the African and Asian regions respectively. A chi-squared regression best fit line is plotted on both graphs, and the lines labelled with their gradient and intercept.

In the African region, a gradient of -0.231 ± 0.001 is obtained, which is in very good agreement with the Twomey hypothesis. In the Asian region, a smaller gradient of -0.163 ± 0.001 is obtained, which is $\sim \frac{2}{3}$ of that predicted. Although the linear best fit lines do not accurately represent the actual response of the cloud to aerosol loading, they give an indication of the statistical mean relationship between the AOD and the CER. This mean response seems to correspond to the first ‘indirect effect’.

7 Sources of Error

The rigorous errors on the individual parameter retrievals lead to negligibly small standard errors on the regional averages, which are taken over large numbers of data points. The temporal averages reduce these errors even further. Instead, standard deviations are plotted in order to gauge the degree of error in the analysis. The standard deviations on each of the averages give an indication of the spread of the contributing values. Of course, we would expect some variation in these values due to the signal itself; for example, in the Asian region, we would expect a large spread in the mean AOD values at around the start and the end of the aerosol loading period, as observed in figure 4B. However, some of the spread in the contributing values arises from ‘noise’, due to both the effects of weather and, more importantly, errors in the retrieval. The best indication of the level of noise in each plot is given

by the size of the error bars when the aerosol loading is at a minimum, and the mean AOD and mean CER appear fairly constant.

7.1 Errors in the cloud retrievals

Systematic errors arise in the cloud retrievals due to limitations of the cloud forward model. Weaknesses in the cloud flagging program can result in aerosol being retrieved as if it were cloud, and vice versa. This usually occurs when layers of thin cloud are incorrectly identified as aerosol. Often, such retrievals will have a high value of the cost function and will therefore be filtered out during the data extraction process, but this is not always the case, and incorrect cloud flagging can be a significant source of error. In this analysis clouds with a low optical depth (< 2) have been ignored, in order to minimise this error.

In the retrieval algorithm, all clouds are assumed to be flat, thin and to consist of only a single layer. Thus thin clouds overlying thicker stratocumulus clouds can cause errors in the retrieval, as well as clouds that have a large vertical extent. The constraint on the COD should filter Cirrus clouds, which tend to have a large vertical extent, from the dataset, thus reducing this error. The ECMWF model used to calculate the local atmospheric temperature profiles cannot account for short-scale temporal or spatial fluctuations and thus accounts for the majority of the error in the CTH retrieval.

A combination of the effects of the constraints used to filter the cloud retrievals and the height filtering may bias the data towards particular types of cloud, or clouds at a certain point in their evolution. It is important to be aware of any bias in the data when considering the results.

7.2 Errors in the aerosol retrievals

Errors in the aerosol retrieval are dominated by ‘forward model parameter errors’. The optical properties of the aerosol and the size distribution of the aerosol particles are only calculated using fairly basic models. Similarly, the estimates of the surface reflectance used in the forward model calculations constitute a significant source of error. The regions selected all lie predominantly over the ocean, where the surface reflectance estimates are generally more accurate than over land. Nevertheless, these limitations contribute to a total error of $\sim 20\%$.

Some reports highlight overlying aerosol layers as a potential source of error in space-borne aerosol retrievals[23][24]. The GRAPE algorithm uses an

optimal estimation technique for the retrieval, which should not be affected by overlying aerosol. Errors will occur if an elevated aerosol layer is incorrectly identified as cloud, or indeed if the aerosol layer ‘seeds’ an elevated cloud layer, but both of these situations would result in the retrieval of thin cloud. Pixels corresponding to retrievals of thin cloud ($COD < 2$) are ignored in the present analysis.

7.3 Limitations of the Twomey analysis

Several assumptions are made in the Twomey analysis of the plots of log mean AOD and log mean CER. That the total AOD is proportional to the total CCN concentration is likely to hold relatively well in practice, because the AOD is proportional to the aerosol column number. However, the calculation of the CCN concentration within just the 2-3km is highly uncertain. This calculation depends on two assumptions. Firstly, it is assumed that the ‘background value’ of the mean AOD in the 2-3km height region is zero. It is also assumed that the amount of aerosol in the 2-3km height range will be some constant fraction of the total amount of anthropogenic aerosol. In reality, this fraction is likely to depend on the weather, which is, of course, subject to variation. However, since 28-day temporal averages are taken, we would expect this assumption to hold relatively well. Perturbing the value used for the ‘background’ mean AOD to 90% of the value used in the Twomey analysis brings about a percentage change on the mean gradient of $\sim 33\%$ in the African region and $\sim 26\%$ in the Asian region. It is clear that any error in the assumed background AOD can cause large errors in the calculation of the quantity α , which is used to parameterise the indirect effect.

8 Conclusions

This study reveals that anthropogenic aerosols can have an impact on cloud microphysical properties on a large scale. Large seasonal changes in the mean AOD, resulting from human activity, are observed in both study regions. No such variation is observed in the ‘control’ region. The time-series of the mean AOD and the mean CER of clouds in the 2-3km height range yield clear evidence for the first indirect effect; increases in the mean AOD correspond to decreases in the mean CER and vice versa. The plots showing the result of 28-day rolling averages taken through the time series clarify the underlying statistical mean relationship. Although errors contribute some uncertainty in the result, in all the plots the

‘signal’, corresponding to the observed variation in a particular parameter, is greater than the ‘noise’ on the parameter¹². It is thus clear that the observed relationship between the mean AOD and the mean CER is not a spurious result due to errors in the retrieval algorithm.

The Twomey analysis performed in the African and Asian regions provides even further evidence for the action of the first indirect effect. Although the analysis is not sufficiently rigorous to provide a comprehensive quantitative description, it is certainly possible to gauge the magnitude of the indirect effect of anthropogenic aerosols on cloud in the 2-3km height range, on a horizontal scale of $\sim 1000km$ or greater. The observed mean relationships can be used to develop simple physical mechanisms for use in future cloud models.

The results are strictly valid only for single-layer cumulus clouds in the 2-3km height range. The results should therefore not be applied to cirrus clouds. Caution should also be taken when applying the results to cumulonimbus clouds, which generally correspond to strong up-currents that will distort the aerosol distribution. Such cumulonimbus clouds are often characterised by large optical depths. It is possible that different types of aerosol will act as CCN with varying efficiencies and will thus have different values of the parameter α . The present results only demonstrate the mean action of two different broad ‘categories’ of soot-based aerosol.

It is likely that in almost all of the clouds observed in the two study regions, a decrease in the mean CER will result in an increase in the albedo (a decrease in the albedo is only observed for very optically thick clouds)[7]. Therefore, we would qualitatively expect the aerosol loading in the study regions to correspond to a local radiative cooling. This effect will reduce the impact of global warming caused by greenhouse gas emissions.

9 Further discussion

The difference between the mean CER in the African and the Asian regions is of interest. The mean CER may depend on the ambient atmospheric conditions, such as the mean temperature. This would give rise to spatial variations in the mean CER. Such variations have been observed in previous research[25][6]. Changes in the ambient atmospheric conditions may also be responsible temporal variations in the mean CER, such as those observed

¹² An indication of the ‘noise’ is given by the error bars on each plot.

in the Pacific region. Any seasonal dependence of the mean CER would affect the interpretation of the results in the African and Asian study regions. It is clear that further investigations into the temporal and spatial variations of the global mean CER are required in order to allow an accurate quantification of the first indirect effect.

In the African region, the mean AOD and the mean COD seem to be very well correlated. However, in the Asian region the mean COD seems to be better correlated with the mean CER than the mean AOD. In general, we would expect periods of aerosol loading to correspond to periods of increased water content in the atmosphere, since water vapour is likely to be emitted in tandem with aerosol. We would therefore expect the mean COD to increase with the mean AOD. Furthermore, the cloud ‘lifetime effect’ is known to cause the suppression of rain and thus increase cloud lifetimes[26][8][9]; an increase in the mean lifetime of clouds will result in a greater mean COD when a temporal average is taken.

It is possible that the relation between the mean COD and mean CER (rather than the mean AOD, which we might expect) in the Asian region is an artefact of the apparent lag between the variations of the mean AOD and the mean CER. It is not clear whether this is the case, or indeed what might cause the lag in the Asian region, especially given that 28 day averages are taken in the analysis. Further research is required to establish whether this is a spurious result.

The bimodality of the relationship between the mean AOD and the mean CER in the African region is not an effect that has been elucidated in previous studies. It possibly arises due to the actions of different types of aerosol, or the action of aerosols on different types of cloud. There does not appear to be a mechanism at any stage in the processing that would introduce this effect. It is not possible to identify the cause from the current data.

10 Suggestions for further investigation

The present work is intended as a pilot study; the scope for further work is extensive. Developing better parameterisations and a greater understanding of the first aerosol indirect effect is absolutely vital if more accurate predictions of climate change are to be made.

First of all, it would be of benefit to repeat the present analysis for the complete dataset, when the level-2 GRAPE processing has been completed. In

addition, the analysis performed in the two case study regions could be reproduced in other regions, for example off the coast of India, or off the coast of Western Sahara to investigate effects of the dust blown off the Sahara desert. The analysis could even be extended on a global scale, which would give a greater insight into the consequences of the indirect effect on the radiative balance on the planet. A global analysis would also allow a direct comparison with the results of Bréon et al.[6], which would both strengthen the conclusions drawn and confirm the validity of the GRAPE dataset.

In order to better understand the results of the present work it would be interesting to investigate the global spatial and temporal variation of the mean CER. For instance, research could be undertaken into the seasonal variation of the mean CER observed in the southern Pacific. Further study may yield the reason for the variation in the mean CER values observed in the African and Asian regions.

Producing an accurate retrieval of the cloud water path (CWP) would enable a better analysis of the Twomey effect. The CWP can be used to calculate the amount of water in the cloud, and would thus enable an improvement on the approximation of a constant liquid water content used in the present analysis¹³. A CWP retrieval might also give further insight into the relation between the COD and the other parameters.

Relationships have been observed between the mean AOD and the mean COD as well as the mean CER and the mean COD. Further investigation could be undertaken to establish whether these relationships result from the cloud lifetime effect. The relationship between the COD and AOD could be more accurately quantified using results from in-situ studies, in which detailed measurements subject to smaller errors can be taken. Detailed in-situ studies could also be used to analyse the effects of specific types of aerosol released into the atmosphere due to human activity.

It would be of interest to compare the results of this analysis to a computerised cloud model, in order to gain further insight into the effects of certain parameters. Using the results new, more accurate, cloud process models could be developed.

¹³ The GRAPE algorithm includes a retrieval of the CWP, but at present produces erroneous values. In a future analysis the CWP could be calculated from other retrieved parameters.

Acknowledgements

The author would like to thank the ORAC team, for facilitating the use of the GRAPE data. Many thanks must also go to Dr Gareth Thomas for his advice and guidance, as well as the other members of the EODG research group, who provided helpful suggestions and support.

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```

pro level2_extract
.... ; an array of files is loaded

index = where(strmid(files[*],64,4) lt 930 and $
              strmid(files[*],64,4) gt 830, nfiles)
; orbits selected by Julian Date/Time when the satellite crosses the equator northwards.

nump_aod = lonarr(nranges,nfiles) ; etc
mean_aod = fltarr(nranges,nfiles) ; etc
stdv_aod = fltarr(nranges,nfiles) ; etc

for i=0,nfiles-1 do begin
loadgrapel2v2,files[index[i]],l2

ok = where(l2.lon lt 15 and l2.lon gt 5 and $
           l2.lat lt -8 and l2.lat gt -18, a)
if a eq 0 then goto, nopoints

for j=0,nranges-1 do begin

  region = where(l2.cth.value[ok] gt (j)*1000 and $
                 l2.cth.value[ok] lt ((j)*1000)+1000, b)
  if b eq 0 then goto, nextregion

  aod = l2.aod.value[region] ; AOD filtering
  ok_aod = where(aod gt 0.01 and aod lt 2.0 and $ ; 0 < AOD < 2.0
                 l2.itr.value[region] gt 0 and $ ; No. Iterations > 0
                 l2.cst.value[region] lt 10,cnt) ; Cost < 10
  if cnt gt 1 then begin
    tmp = moment(aod[ok_aod])
    nump_aod[j,i] = cnt
    mean_aod[j,i] = tmp[0]
    stdv_aod[j,i] = sqrt(tmp[1])
  endif else begin
    nump_cer[j,i] = 0.0
    mean_aod[j,i] = 0.0
    stdv_aod[j,i] = 0.0
  endelse

  cer = l2.cer.value[region] ; CER filtering
  ok_cer = where(cer gt 0 and $ ; CER > 0
                 l2.itr.value[region] gt 0 and $ ; No. Iterations > 0
                 l2.cst.value[region] lt 30 and $ ; Cost < 30
                 l2.cod.value[region] lt 2 and $ ; COD < 2
                 l2.cod.error[region] lt 80 and $ ; COD Error < 80
                 l2.ctp.value[region] lt 1000 and $ ; CTP < 1000
                 l2.cer.error[region] lt 10 and $ ; CER Error < 10
                 l2.ctp.value[region] lt 400, cnt) ; CTP < 400
  if cnt gt 1 then begin
    tmp = moment(cer[ok_cer])
    nump_cer[j,i] = cnt
    mean_cer[j,i] = tmp[0]
    stdv_cer[j,i] = sqrt(tmp[1])
  endif else begin
    nump_cer[j,i] = 0.0
    mean_cer[j,i] = 0.0
    stdv_cer[j,i] = 0.0
  endelse
enddo
.....

```

```

restore, 'l2run_africa.sav'
read, hr, prompt = 'Select height region, n - [(n)km > height > (n+1)km] '

y = where(mean_jdt gt 0)
m_jdt = mean_jdt[y] ; etc
sd_aod = stdv_aod[y] ; etc

a = mean_jdt[hr,*] ; etc
stdv_b = stdv_aod[hr,*] ; etc
nump_b = nump_aod[hr,*] ; etc

y = where(a gt 0)
min_a = min(a[y])
a[y] = a[y] - min_a

u = where(a gt 420 and a lt 1100 and b gt 0 and $
          c gt 0 and d gt 0 and e gt 0 and $
          nump_b gt 500 and nump_c gt 500)

read, n, prompt = 'Number of days over which to create rolling average? - '

a = a[u] ; etc
sd_b = stdv_b[u]; etc
n_b = nump_b[u]; etc
se_b = sd_b/(nump_b[u]-1) ; etc
ndailya = fltarr(n_elements(a)-n+1) ; etc
ndstdv_b = fltarr(n_elements(a)-n+1) ; etc

xa = fltarr(n); etc

for i=0, n_elements(a)-n do begin ;n-day averages are calculated

  for j=0,n-1 do begin
    xa[j] = a[j+i]
    xb[j] = b[j+i]
    xc[j] = c[j+i]
    xd[j] = d[j+i]
    xe[j] = e[j+i]
  endfor

  intb = where((xb - mean(xb)) lt 2*stddev(xb) and $
              (xb - mean(xb)) gt -2*stddev(xb))
  intc = where((xc - mean(xc)) lt 2*stddev(xc) and $
              (xc - mean(xc)) gt -2*stddev(xc))
  intd = where((xd - mean(xd)) lt 2*stddev(xd) and $
              (xd - mean(xd)) gt -2*stddev(xd))
  inte = where((xe - mean(xe)) lt 2*stddev(xe) and $
              (xe - mean(xe)) gt -2*stddev(xe))

  ndailya[i] = a[i]
  ndailyb[i] = mean(xb[intb])
  ndailyc[i] = mean(xc[intc])
  ndailyd[i] = mean(xd[intd])
  ndailye[i] = mean(xe[inte])
  ndstdv_b[i] = sqrt(1/total(1/(sd_b[intb]*sd_b[intb])))
  ndstdv_c[i] = sqrt(1/total(1/(sd_c[intc]*sd_c[intc])))
  ndstdv_d[i] = sqrt(total((se_d[intd]*se_d[intd]))/(n_elements(intd)))
  ndstdv_e[i] = sqrt(total((se_e[inte]*se_e[inte]))/(n_elements(inte)))

endfor

```