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Investigation into clouds and precipitation over an urban area using micro rain radars, satellite remote sensing and fluorescence spectrophotometry

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A B S T R A C T

The observation and modeling of the indirect effects of aerosols on clouds remain an enormous challenge. Aerosols have a significant yet complicated impact on the precipitation processes. They can either enhance or suppress precipitation depending upon type of aerosol, seasonality, climate regime, cloud type or orographic profile of a region, particularly over populated areas. In order to observe and examine both cloud and precipitation processes, a combination of both satellite and ground-based remote sensing techniques can be employed. This paper presents the results from three years of data collection in Birmingham, United Kingdom. It describes and explains the application of a range of complimentary techniques: fluorescence spectrophotometry to examine dissolved organic carbon compounds in rainwater samples; satellite analysis tools are used to assess cloud-top microphysics; and an array of vertically-pointing micro-rain radars (MRRs) are used to assess variations in drop size distribution (DSD) for categorized events. Events are classified as microphysically 'maritime' or 'continental', showing that full development of the ice phase was reached at relatively warm temperatures for microphysically 'maritime' events, but at colder temperatures for microphysically 'continental' events. The importance of updrafts in severe thunderstorms and tornadic events is highlighted. High rainwater content of tyrosine-like substances (TYLIS) and tryptophan-like substances (TRYLIS) is found to be associated mainly with microphysically 'maritime' events, providing evidence for these substances acting as ice nuclei at relatively warm temperatures. High rainwater content of humic-like substances (HULIS) is associated with both microphysically 'maritime' and 'continental' events due to the complexity of such substances. As might be expected, continentally-sourced events had a similar structure to microphysically 'continental' events, whereas maritime-sourced events differed in their microphysical structure, indicating the local impacts on their microstructure. The DSD appears to vary between different events — for example, continentally-sourced, microphysically 'continental', convective events with low rainwater TRYLIS have a DSD containing fewer smaller droplets, whereas maritime-sourced, microphysically 'maritime', stratiform events with high TRYLIS had a DSD containing a greater number of smaller droplets. Satellite observations and vertically-pointing radars were found to be useful for analyzing clouds and precipitation since they provide a wealth of information to allow microphysical parameters to be investigated in detail.

1. Introduction

Aerosols play important roles in the atmosphere, having both direct and indirect effects on climate. The direct aerosol effect is caused by the absorption and scattering of solar radiation, whereas the indirect effect is linked to the action of aerosols as cloud condensation nuclei (CCN), thereby affecting the initial cloud droplet number concentration, albedo,

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precipitation formation, and lifetime of warm clouds (Pruppacher and Klett, 2000). Evaluating the indirect effects of aerosol on clouds remains an enormous challenge from both the observational and modelling perspectives. This has prompted atmospheric scientists to explore the impact of increasing anthropogenic activities on cloud and precipitation processes.

It is recognised that different aerosols have varying impacts upon precipitation. Theories of precipitation suppression caused by an increase in small CCN have been proposed by Twomey (1974) and Rosenfeld (2000). Small aerosols cause a narrowing of the size distribution of cloud droplets that lead to reduced or suppressed precipitation, since a range of droplet sizes are required for warm rain to develop. Therefore, in polluted clouds it is suggested that there are too many small droplets and too few larger or 'giant' droplets for efficient precipitation to occur. However, it has also been suggested that increasing CCN concentrations prolongs the lifetime of clouds and ultimately acts to increase precipitation (Shepherd, 2005). Due to reduced collection efficiency, such clouds may continue to ascend to altitudes where graupel and ice crystals form — these clouds are deeper and produce heavy rain, lightning and hail. Therefore, under certain conditions, a delay in the onset of warm rain due to aerosols can result in delayed downdraft formation, allowing for more invigorated updrafts which produce deeper and stronger convection. This effect may be experienced 'downwind' of urbanised regions. Furthermore, the presence of 'giant' nuclei can also act to enhance precipitation. Giant aerosols produce large cloud droplets near the cloud base — the effects of such giant CCN are significant when the concentration of small, Aitken nuclei is high, as in urban clouds. Giant CCN also act as a destabilising factor, by accelerating collisions and coalescence between the water drops which causes early development of large drops in lower parts of the cloud. Giant CCN accelerate precipitation formation through the ice phase, due to formation of ice by nucleation. Large droplets formed by the giant CCN produce graupel particles earlier — these have high coagulation efficiency with drops and therefore grow more rapidly as they are lifted in the updraft region, yet they remain close to the cloud base which can also promote ice multiplication processes in supercooled regions (Yin et al., 2000). Aerosols therefore have a significant, yet complicated impact on the precipitation process: they can either enhance or suppress precipitation depending upon type of aerosol, seasonality, climate regime, cloud type or orographic profile of a region, particularly over populated areas. It is also important to note that, in addition to aerosols, there are also other factors which have an impact on precipitation over urban areas, including thermodynamic effects, (Shepherd et al., 2002; Shepherd and Burian, 2003), mechanical effects (Bornstein and Lin, 2000), and bifurcation caused by urban canopies (Loose and Bornstein, 1977; Bornstein and Lin, 2000).

Water-soluble organic compounds (WSOC) are particularly important in heterogeneous nucleation and the formation of clouds and precipitation. A large proportion of rainwater WSOC is still uncharacterized — little is known about the chemical compounds present, their sources, temporal and spatial patterns of variation, and the subsequent impact on climate and the environment (Muller at al., 2008). WSOCs can influence cloud albedo, increase cloud condensation nuclei (CCN) concentrations, contribute to rainwater pH, visibility impairment and photochemical processes, and is a nutrient input to rivers and ecosystems. WSOC can be formed via physical and chemical aging. Sources of WSOC compounds in rain include: primary anthropogenic emissions (motor vehicle exhaust, tyre and asphalt wear, cooking), primary biogenic emissions (biomass burning, soil-derived humic matter resulting from combustion, thermal breakdown of plant ligins and cellulose), marine sources (bubble bursting on ocean surfaces), and secondary organic aerosol formation mechanisms (condensation, evaporation, photochemical reactions, oligomerization and aerosol-phase polymerization). Since a large proportion of WSOC is incorporated into rain droplets within the cloud environment (Barth et al., 2001), analysing rainwater concentrations of such compounds can provide an indication of atmospheric concentrations within the cloud environment. This can then be used to examine the atmospheric conditions under which the precipitation developed — *in situ* data collection at such altitudes would only be possible from balloon or aircraft platforms during the observed event, whilst satellite data would not provide the temporal and spatial resolution required for an investigation into atmospheric conditions during individual precipitation events. Despite the lack of a universal standard for accurate calibration (Muller et al., 2008), rainwater fluorescence intensity can be used as a proxy for the concentrations of substances in the rain samples. This technique provides a non-invasive, rapid method to examine the content of precipitation samples. Humic-like substances (HULIS), tyrosine-like substances (TYLIS) and tryptophan-like substances (TRYLIS) were identified in the rainwater samples (Muller et al., 2008). Atmospheric HULIS are high molecular weight compounds which are similar to terrestrial and aquatic humic substances, but do have substantial differences, such as smaller *average* molecular weight, lower aromatic moiety content, weaker acidic nature, a higher aliphatic component and better surface activity and thus better droplet activation ability compared to terrestrial and aquatic humic substances, due to greater number of solute species in HULIS (Graber and Rudich, 2006). TYLIS and TRYLIS are amino acids: proteins and biogenic matter have recently been found to be effective ice nuclei, especially at relatively warm temperatures (Christner et al., 2008).

In order to observe and examine both cloud and precipitation processes, a combination of satellite and ground-based remote-sensing techniques can be employed. Satellite remote-sensing techniques are increasingly used to examine clouds since aircraft observational tools cannot characterise the true evolution of cloud microphysical, spatial and temporal structure in the cloud droplet scale and relate this to properties of CCN or other environmental factors (Martins et al., 2007). Therefore satellite observations tools, such as temperature-effective radius (T–r_e) relationships and Cloud-Aerosol-Precipitation Satellite Analysis Tool (CAPSAT), are necessary. CAPSAT was developed by Lensky and Rosenfeld (2008) for cloud microphysical characterisation and aerosol–cloud interaction detection. It uses red–green–blue (RGB) composites of selected multispectral channel observations to represent much of the physical information retrieved by the observations from the Meteosat Second Generation (MSG) satellite (Lensky and Rosenfeld, 2008). Combinations of the selected channels can be used to qualitatively examine precipitation forming processes, for example, cloud drop size, which is a major factor in cloud microstructure that determines their precipitation forming processes.

Furthermore, the microphysical evolution of convective cloud elements can be examined using T–r_e relationships. This is represented by the composition of the instantaneous values of the tops of clouds at different heights, based on the knowledge that cloud droplets form mainly at the base of convective clouds and grow with increasing height (decreasing temperature). T can be related to r_e in a given cloud cluster, to provide information regarding cloud microstructure and precipitation processes (Rosenfeld et al., 2008). Areas with smaller r_e for a given T are associated with younger cloud elements, whereas larger r_e for the same T are associated with the more mature cloud elements, such that droplet growth by coalescence has progressed and ice crystals have had time to develop (Rosenfeld et al., 2007). Clouds are considered microphysically 'maritime' when the concentration of cloud droplets at the base of the cloud is less than 100 droplets cm^{-3}, whilst clouds are considered microphysically 'continental' when the cloud droplet concentration at cloud base is approximately 1100 droplets cm^{-3} (Rosenfeld and Woodley, 2003). The difference between continental and maritime clouds means that precipitation from continental clouds forms mainly from ice processes, whilst it is mainly warm rain processes that occur in maritime clouds — even in very deep maritime convective clouds which extend into the sub-freezing temperatures (Rosenfeld and Ulbrich, 2003).

Conventional precipitation measurement techniques rely upon gauge measurements, including tipping-bucket rain gauges, augmented through the development of distrometers, ground-based radars, and satellite instrumentation. More recently, vertically-pointing radars have been introduced study high-resolution sampling of precipitation events (Fabry and Zawadzki, 2000). Vertically-pointing micro rain radars (MRRs) provide a new method of monitoring rainfall, allowing a more precise rain-rate estimate (Deiderich et al., 2004). MRRs are small, with a 0.5 m efficient aperture diameter dish, have low power consumption and can be attached to a laptop for operation. They measure the backscatter of radiation from precipitation-sized particles from the surface up to 6000 m, enabling a number of parameters to be generated including the fall velocity (W), drop size distribution (DSD) — which varies greatly with precipitation type and rate — liquid water content (LWC), rain rate (R) and reflectivity (Z), allowing for the microstructure of the precipitation to be analysed. Fig. 1 shows an example of the vertical profile of a number of parameters for a typical precipitation event. MRRs, operating at 24 GHz measure not only the backscattered radar signal, but also the Doppler shift of the return signal, which allows a measure of the falling velocity of the rain drops to be made. Rain drops of different diameters have different falling velocities and thus the DSD can be resolved and used to determine rain rate as an alternative method to the usual Z–R relation of weather radars. MRRs are sensitive to low rain rates allowing 0.1 mm/h in 10 second averaging period to be measured compared to tipping bucket rain gauges with a quantitative resolution of 0.1 mm per tip. The DSD (expressed as the number of drops per cubic meter of air per millimeter diameter, $m^{-3}\ mm^{-1}$), is dependent upon the precipitation processes during raindrop formation, growth, transformation and decay occurring on a microphysical scale within the cloud environment (Roy et al., 2005). Rosenfeld and Ulbrich (2003) summarise the impacts of these processes as condensation growth, evaporation, break-up, accretion, collision and coalescence, further affected by updrafts and downdrafts that modify the DSD. Few studies have compared the DSD curves for different events at the same locations. However, one study did carry out research at sites within the same region: Rosenfeld and Ulbrich (2003) compared DSD at paired locations (one continental and one maritime) in the same region, but at different times in order to assess the differences between maritime and continental cloud microstructure. The continental clouds contained a far greater concentration of larger droplets and a smaller concentration of small droplets. It was suggested that the main causes for these differences included the extent of coalescence, warm versus cold precipitation processes, strength of updrafts, and evaporation.

This paper presents results from three years of data collected in Birmingham, United Kingdom using a number of complimentary techniques. Satellite data and radar data will be used to investigate the observations of cloud-top and drop-size distribution dependency upon source area, storm type, microphysical classification and rainwater florescence. The techniques used are discussed and the results are examined in detail: Firstly, trends in cloud microphysics are examined using a classification scheme applied to each event — a select number of case studies are also examined in more detail to investigate 'extreme' events. The corresponding rainwater fluorescence values and source areas are also investigated for each microphysical classification. Secondly, events are classified according to their precipitation microstructure (using the DSD), and the corresponding source area, storm type and fluorescence value is examined for each class. Finally, conclusions are drawn and recommendations suggested.

2. Method

Satellite and ground-based data were collected and used, along with additional meteorological data, to assess the variations in clouds and precipitation occurring during individual precipitation events over the urban area of Birmingham, United Kingdom (Fig. 2). A number of events have been selected for inclusion in this study. These range from typical mid-latitude precipitation regimes, to data from a tornadic event which occurred in July 2005. The use of fluorescence spectrophotometry to examine dissolved organic carbon compounds in rainwater samples is demonstrated. Additional information from CAPSAT image analysis tool and T–r_e plots are used to assess cloud-top microphysics information, and data from an array of vertically-pointing micro-rain radars are used to assess variations in DSD for categorized events.

2.1. Cloud microphysics

Cloud top microphysical information was obtained from observations made by the SEVIRI instrument on board the EUMETSAT Meteosat Second Generation (MSG) satellite at a temporal resolution of 15 min covering the duration of each precipitation event — therefore the satellite data provides a broad representation of the cloud structure during each precipitation event. The image analysis tool, CAPSAT, was used to qualitatively examine cloud microphysics and aid identification of convective clouds over Birmingham using a number of standard RGB compositions (see Lensky and Rosenfeld, 2008 for details). Raw data was also extracted for a 3°×3° latitude/longitude area covering Birmingham to quantitatively investigate the cloud droplet effective radius (r_e) and cloud-top temperatures (T). Droplet r_e is proportional to the sum of the cloud droplet volumes divided by the sum of the surface areas of the droplets in the measurement area. During daylight, the large absorption in the 1.6 and 3.9 μm bands compared to the non-absorbing visible channel makes it possible to retrieve r_e. Cloud-top temperatures were retrieved from the SEVIRI thermal channels, allowing T–r_e plots to be generated to examine the precipitation forming processes and the microphysical state of each event. The microphysical structure for each event was based on the classification schemes of the convective clouds according to the shape of the T–r_e relations (the two extremes being "continental" and "maritime") from Rosenfeld and Woodley (2003) and Rosenfeld et al. (2008).

2.2. Precipitation microstructure

The MRRs retrieved parameters, W, DSD, LWC, R and Z, were used to help examine and identify prevailing storm type, and examine precipitation microstructure. Parameters were measured every 10 s and averaged to provide 60-second samples of each parameter for every 200 m from the surface up to 6000 m. During the initial comparison period it was found that the lowest height bin underestimated precipitation, possibly due to ground level turbulence, therefore data from the second height bin was used for analysing ground level precipitation. DSD retrievals are dependent upon radar calibration constant C. This was determined by comparison

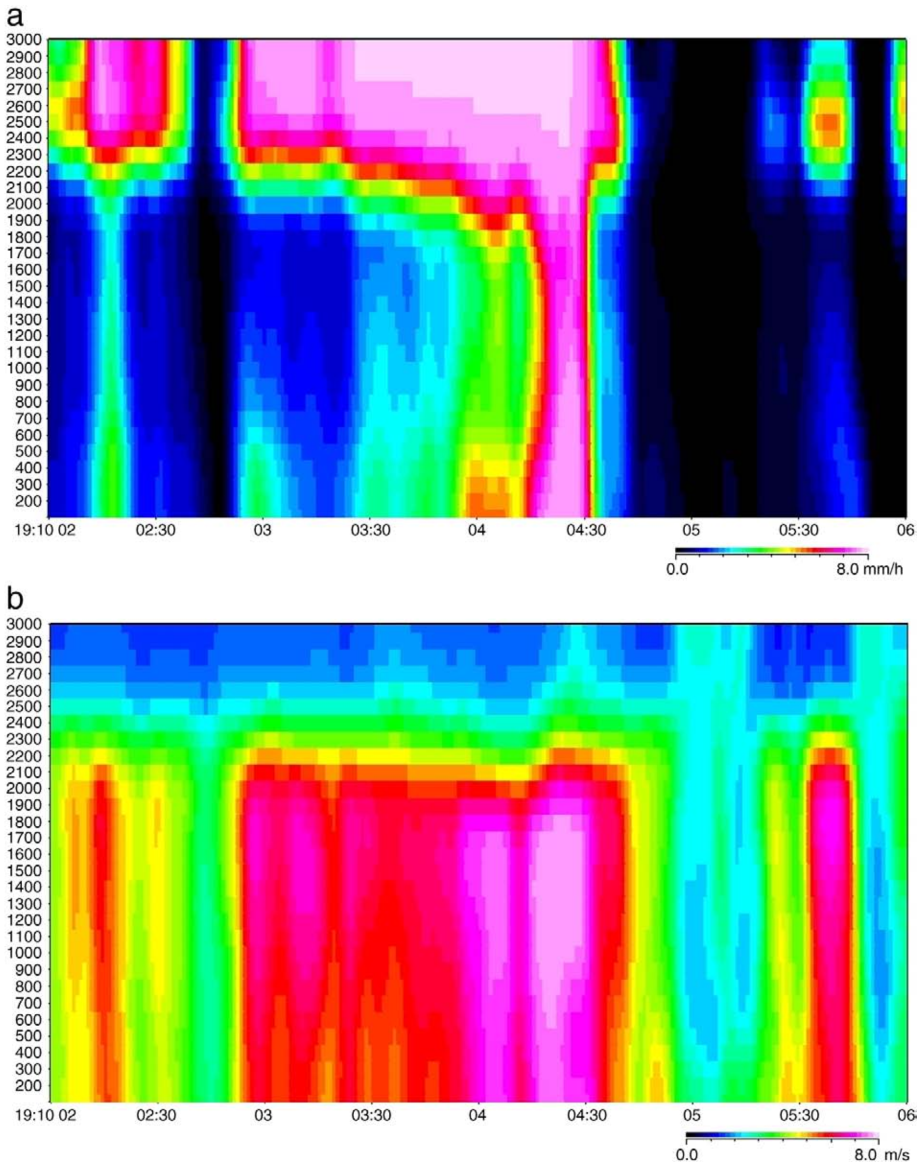

Fig. 1. MRR vertical profiles of a) R; b) W; c) Z and d) LWC on 19/10/06 for a cold front event. x-axis: time (hh:mm), y-axis: height (m).

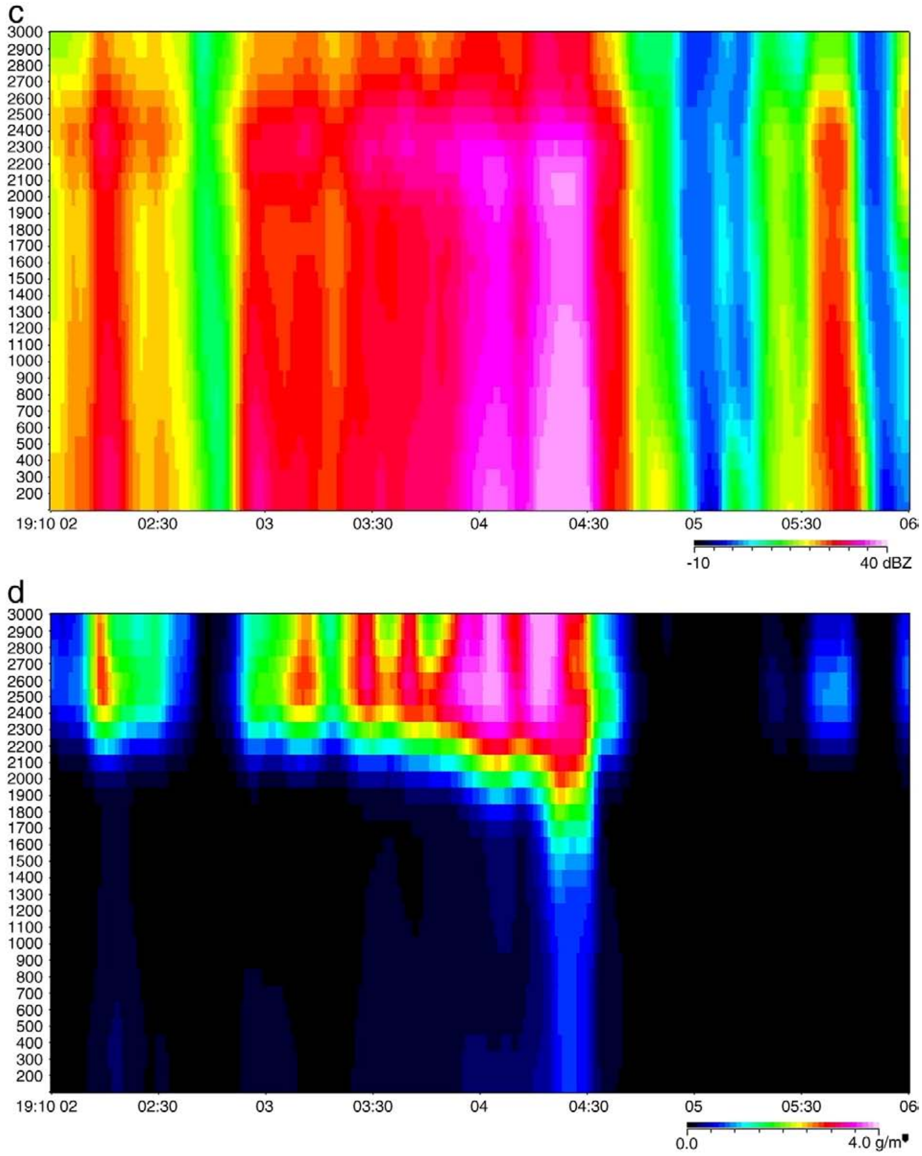

Fig. 1 (*continued*).

with in situ rain rate measurements in selected environmental conditions, and its uncertainty is estimated to be approximately ±10% by the manufacturers (METEK, 2005).

2.3. Rainwater fluorescence

Samples from individual precipitation events were collected in Birmingham between April 2005 and January 2008. These were analysed using fluorescence spectrophotometry in order to assess rainwater HULIS, TYLIS and TRYLIS content. A more detailed overview of these fluorophores, the sampling technique used, associated uncertainties and results from the rainwater fluorescence analysis of samples used in this study, can be found in Muller et al. (2008). Precision (one standard deviation) of triplicate fluorescence data sets was ±3%. This fluorescence data, along with other meteorological variables, including particulate matter concentrations, wind speed, wind direction, back trajectory analysis, and synoptic charts, were used to categorise and highlight precipitation events of interest.

2.4. Additional data

The Hybrid Single-Particle Langrangian Integrated Trajectory model (HYPLIT) was used to compute a history of the air movement prior to the sampling period. The model provides trajectories using accurate synoptic scale wind data generated from numerical weather prediction models. The length of time a back-trajectory is calculated is important: if a trajectory is too long then more than one evaporation–precipitation cycle may occur (James et al., 2004). Five-day back-trajectories starting at 10 m, 100 m, 500 m, 1000 m, and 1500 m (~850 hPa) levels were calculated, since they represent surface levels, and

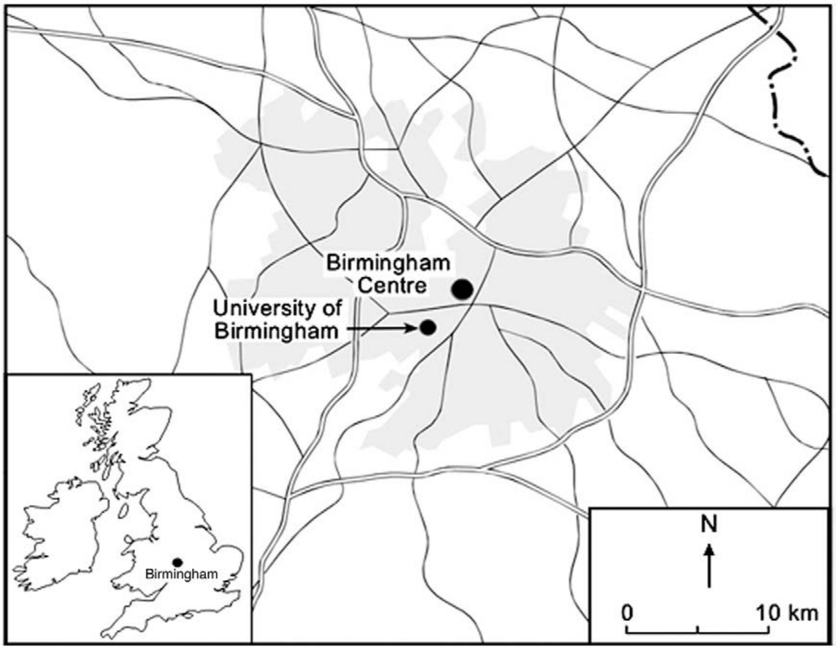

Fig. 2. Map of the location of Birmingham within West Midlands conurbation (shaded) and the UK (inset), and the location of the sampling site (University of Birmingham).

approximate upper limits of the boundary layer by night and day respectively (Witt et al., 2007; Wayne, 1999). These were used to identify the source area and the air mass associated with each event. Furthermore, synoptic charts at 0000 h and 1200 h were obtained for all the sampled events, along with descriptions of the meteorological conditions covering the periods to provide an overview of the prevailing conditions, to help identify rainfall type, and to aid the data analyses. Fluorescence data, personal weather logs, meteorological data, synoptic charts and back trajectories were used to classify each event — Fig. 3 shows a typical back-trajectory for 'polluted' and 'clean' events.

When analysing the results, it is important to acknowledge the uncertainties associated with a study involving a range of techniques — for example: instrument error and calibration uncertainties; the robustness of the classification scheme used to determining convective and stratiform precipitation; the differences in sampling times between the various instruments; the representativeness of the rainwater florescence analysis for determining upper-level conditions since it is not possible to determine the amount of WSOC incorporated within and below the cloud base.

3. Results and discussion

3.1. Cloud microphysics

3.1.1. Classification

Fig. 4 shows the cases which had a microphysically 'maritime' state (from highly maritime to moderately maritime). The maritime clouds start with relatively large r_e at their base, reaching and exceeding the precipitation threshold of 14 μm a short distance above the base, suggesting rain formed by droplet coalescence which in many cases starts at this height (Rosenfeld and Lensky, 1998). There is also evidence of a diffusional droplet growth zone in many of the cases since the clouds originate from clean, maritime environments (Martins et al., 2007). Microphysically 'maritime' clouds are typically associated with the presence of a 'rainout' zone. A deep 'rainout' zone indicates fully developed warm rain processes in the maritime clouds.

Further increases in r_e at supercooled levels (<0 °C) indicates the formation of the ice phase where large droplets freeze, resulting in a shallow mixed-phase zone. The maximum value of r_e is reached at relatively warm temperatures (in one case −12 °C). The point at which maximum r_e is reached, indicates full development of the ice phase at that temperature. Rosenfeld and Lensky (1998) found that the full development of the ice phase can reach temperatures as high as −7 °C. The observations of warm glaciation temperatures in microphysically 'maritime' clouds are consistent with numerous aircraft observations of tropical maritime clouds with almost complete glaciation (Jorgensen and LeMone, 1989). The glaciation temperatures found in this study are therefore slightly colder than previous observations, indicating that these 'maritime' events are not 'extreme' cases.

Fig. 5 shows the T–r_e plot for a number of cases associated with clouds that are microphysically 'continental': these clouds are composed of many small droplets. It is important to note that all of these plots are for precipitation events despite the scarcity of a 'rain out' zone. Small droplets, r_e <14 μm, are observed approaching and in some cases below a temperature of −10 °C. This indicates droplet growth mainly by diffusional processes with little coalescence. Maximum r_e is reached between −22 °C and −38 °C, which implies full development of the ice phase between these temperatures. Therefore, precipitation is not formed as a result of coalescence but rather in the ice phase as hail, graupel or snow in the deep mixed phase zone.

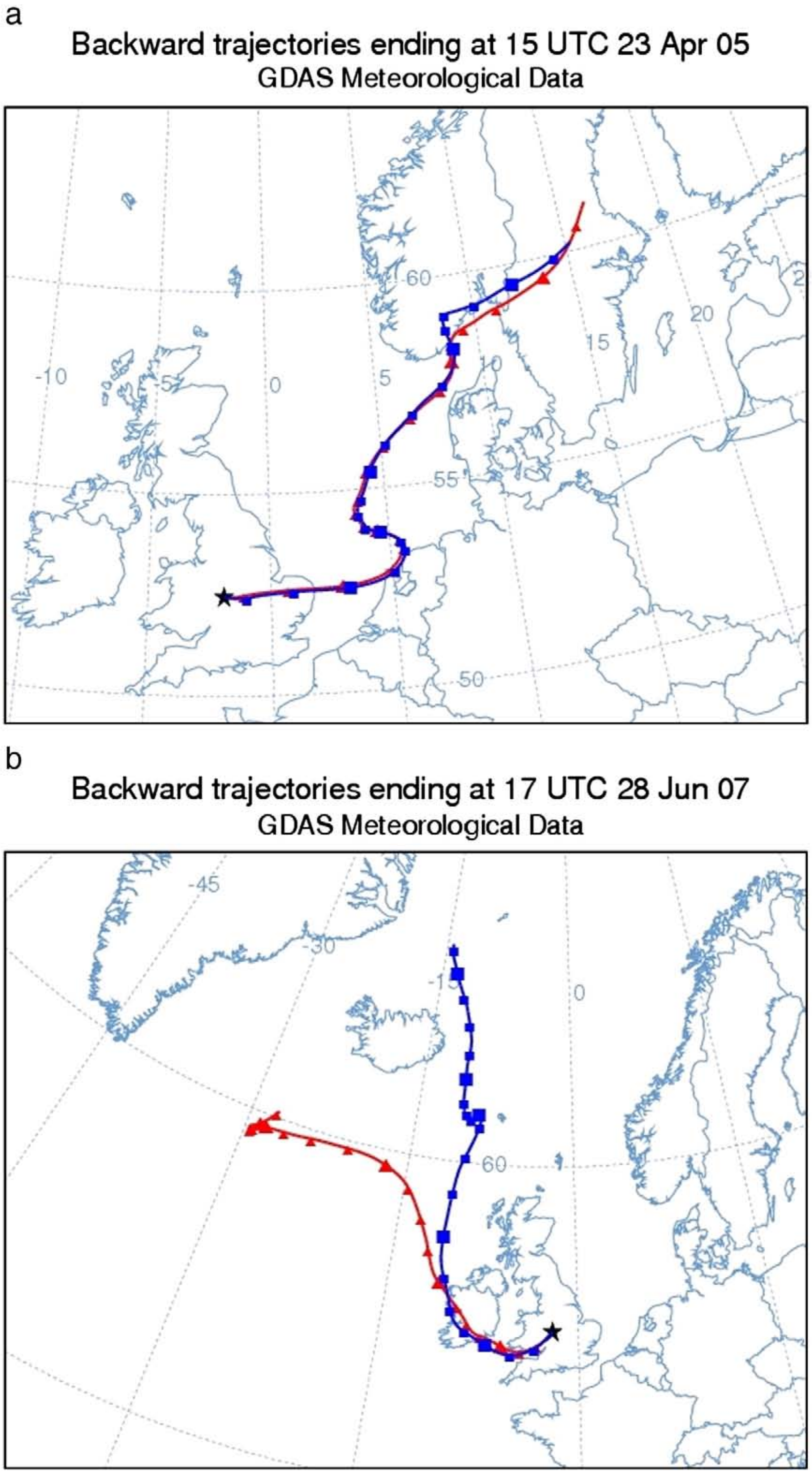

Fig. 3. A 'typical' NOAA HYSPLIT back-trajectory at 500 m (red) and 1000 m (blue) for a) a 'polluted' event (23/04/05) and b) a 'clean' event (28/06/07).

3.1.1.1. Case studies: tornadic event and severe thunderstorms. Thunderstorms typically develop as a result of increased convection in an unstable, humid atmosphere where air temperature decreases with height, resulting in large droplets developing as a result of vigorous updrafts. Thunder and lightning occurred during a number of the sampled events.

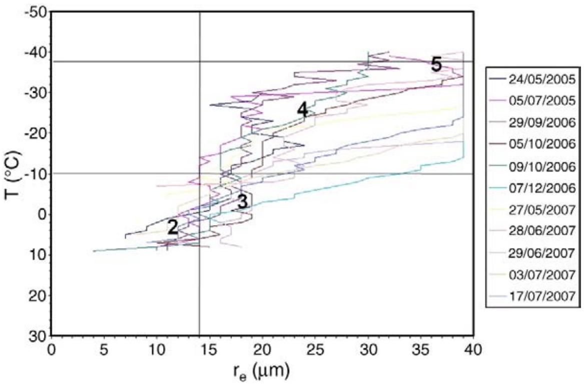

Fig. 4. T–r_e relationships for a 3°×3° area surrounding Birmingham for a number of events showing microphysically maritime characteristics. Plotted are the event medians. For reference: the vertical line indicates the 14 µm precipitation threshold; the horizontal line at −10 °C indicate arbitrary glaciation temperature, whilst the line at −38 °C indicates the homogeneous freezing isotherm. Numbers represent the approximate microphysical zones base on: 2 = Droplet coalescence growth zone; 3 = Rainout zone; 4 = Mixed phase zone; 5 = Glaciated zone.

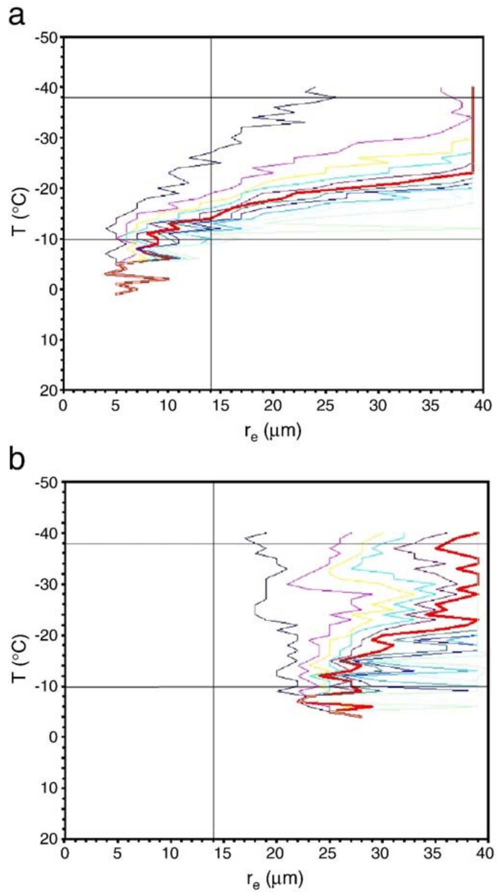

Fig. 6. T–r_e relationships for a 3°×3° area surrounding Birmingham for a number of events: a) 24/06/05 (continental, 'clean') and b) 24/10/05 (maritime, 'polluted'). Plotted are the percentiles of the r_e for each 1 °C interval (these represents younger and more mature cloud elements). The thick red line indicates the median. For reference: the vertical line indicates the 14 µm precipitation threshold; the horizontal line at −10 °C indicate arbitrary glaciation temperature, whilst the line at −38 °C indicates the homogeneous freezing isotherm.

Fig. 6 shows two such events, although both have very different T–r_e relationships. Fig. 6(a) shows a microphysically 'continental' profile, whilst the profile in Fig. 6(b) indicates a more microphysically 'maritime' structure. These events are examined in order to investigate dynamical impacts on T–r_e relationships. Stronger updrafts delay the growth of r_e to greater heights (lower T) since there is less time for cloud and raindrops to grow by coalescence (Rosenfeld et al., 2008). These strong updrafts also postpone the development of a mixed phase zone and glaciation to colder temperatures. Fig. 7 shows the 'convective storm' RGB composite for these events. In (a), cloud tops colder than −38 °C are identified as orange shades and are composed of small ice particles, likely to have formed by homogeneously glaciated cloud water. These may have ascended with strong updrafts and formed the tops of the coldest clouds. In (b), large ice particles formed by heterogeneous glaciation appear pink and occur at cloud tops warmer than −38 °C. Brighter 3.9 µm reflectances can therefore be an indicator of strong updrafts (Rosenfeld et al., 2008). One tornadic event was sampled on 28th July 2005: the rain samples had high fluorescence values simultaneous with high ambient particulate matter concentrations. The T–r_e curve shown in Fig. 8 is for SEVIRI data retrieved just prior to the tornado touching down over Birmingham. The curve shows that the clouds contained smaller droplets and that the onset of precipitation (r_e > 14 µm) occurred at a colder temperature, indicated by the lack of a 'rainout' zone and the presence of a large 'mixed phase' zone.

There appears to be no 'typical' T–r_e plot for occasions when thunderstorms or tornadoes occur — the examples above show two extreme cases under different situations. Williams et al. (1992) noted that more frequent lightning occurred in 'continental' clouds compared to maritime clouds, suggesting that this was due to the fast conversion of cloud water to precipitation in 'maritime' clouds which caused convective

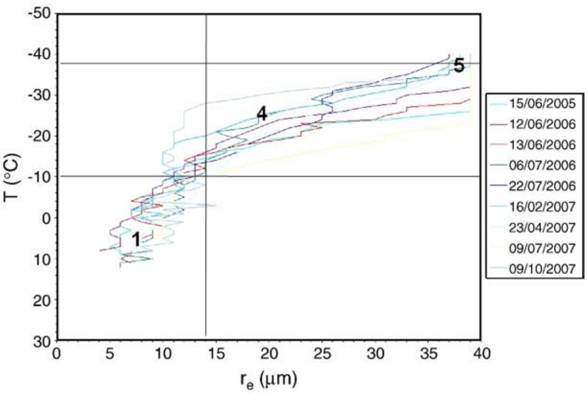

Fig. 5. T–r_e relationships for a 3°×3° area surrounding UoB for a number of events showing microphysically continental characteristics. Plotted are the event medians. For reference: the vertical line indicates the 14 µm precipitation threshold; the horizontal line at −10 °C indicate arbitrary glaciation temperature, whilst the line at −38 °C indicates the homogeneous freezing isotherm. Numbers represent the approximate microphysical zones: 1 = Diffusional droplet growth zone; 4 = Mixed phase zone; 5 = Glaciated zone.

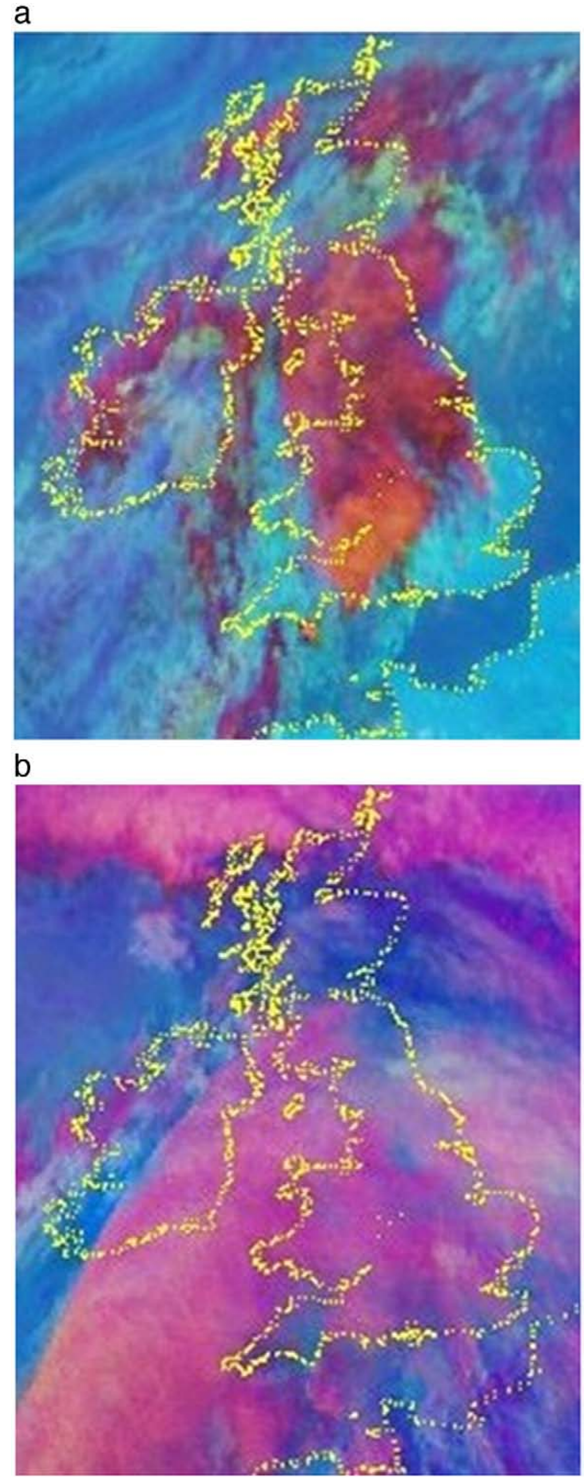

Fig. 7. CAPSAT image of Cb microstructure a) 24/06/05 (continental, 'clean') and b) 24/10/05 (maritime, 'polluted').

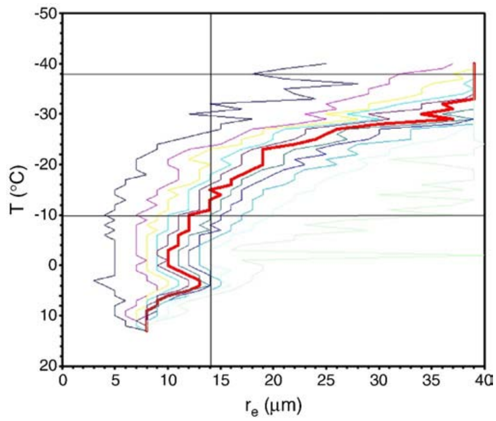

Fig. 8. T–r_e relationship for 28/07/05 for a 3°×3° area surrounding Birmingham. Plotted are the percentiles of the r_e for each 1 °C interval (these represents younger and more mature cloud elements). The thick red line indicates the median. For reference: the vertical line indicates the 14 μm precipitation threshold; the horizontal line at −10 °C indicate arbitrary glaciation temperature, whilst the line at −38 °C indicates the homogeneous freezing isotherm.

elements to lose water to precipitation while growing. This leaves less water to be carried aloft to the supercooled zone, and therefore weaker ice precipitation develops. Furthermore, Rosenfeld and Woodley (2003) reported that microphysically 'maritime' clouds show little lightning activity, further suggesting that pollution can act to enhance the occurrence of lightning. However, Williams et al. (1992) recognised that frequent lightning also occurred in clean air during high atmospheric instability, suggesting this was due to strong updrafts leaving little time for the formation of warm rain, due to large raindrops carried up to the supercooled zone where they freeze and take part in the cloud electrification process. The events above cover both 'polluted' and 'clean' events, and display both microphysically 'maritime' and 'continental' profiles, indicating the importance of strong updrafts in the development of thunderstorms and tornado development. Rosenfeld et al. (2008) suggested the use of T–r_e signatures in clouds ahead (in terms of space and time) of hail and tornadic storms can be used to predict their occurrence at substantial lead times of up to 2 h. It was found that greater updraft manifested in T–r_e plots when: (a) glaciation temperature is reached at a lower temperature, (b) a linear T–r_e line occurs for a larger temperature interval and, (c) the r_e of the cloud at its glaciation temperature is smaller. This can be used to determine clouds with strong updrafts to possess a risk of large hail and tornadoes. For example, colder glaciation temperatures and smaller r_e at cloud base indicate higher probability for a tornadic event.

It is evident that the two thunderstorm events are most similar to the continental case with moderate updrafts and the 'maritime' case with moderate updrafts, whilst the tornadic event is also most similar to the 'continental' case with moderate updrafts. This indicates that updraft speed is extremely important to the development of extreme convective storms, perhaps even more so than microphysical structure impacting on the severity and/or duration of the storm.

3.1.2. Fluorescence

T–r_e plots were generated for the most extreme cases: 'polluted' events with high rainwater TYLIS, TRYLIS, HULIS fluorescence, and 'clean' events with low rainwater TYLIS, TRYLIS, HULIS fluorescence. Although the classification was based on rainwater collected at the University of Birmingham, it is likely to be representative of the rainfall over the area since rainwater fluorescence appears to be related to the prevailing air mass and source area (Muller et al., 2008). Fig. 9 shows the plots for all the selected cases (summarised in Table 1). In general, events with high rainwater fluorescence appear to be associated with more microphysically ‘maritime’ clouds, whilst low fluorescence events are associated with microphysically ‘continental’ or ‘transitional’ clouds, which is perhaps unexpected, since ‘continental’ clouds correspond to a larger number of cloud condensation nuclei, indicative of polluted clouds.

Rainwater TYLIS and TRYLS in particular, appear to be highest during events with both microphysically ‘maritime’ clouds *and* those associated with a maritime-source area. Christner et al. (2008) showed that the activity of most known biological ice nuclei is mediated by proteins or proteinaceous compounds. Ice nuclei are transported from long distances and maintain ice nucleating ability in the atmosphere and play an important role in the initiation of ice formation, especially when minimum temperatures are warm.

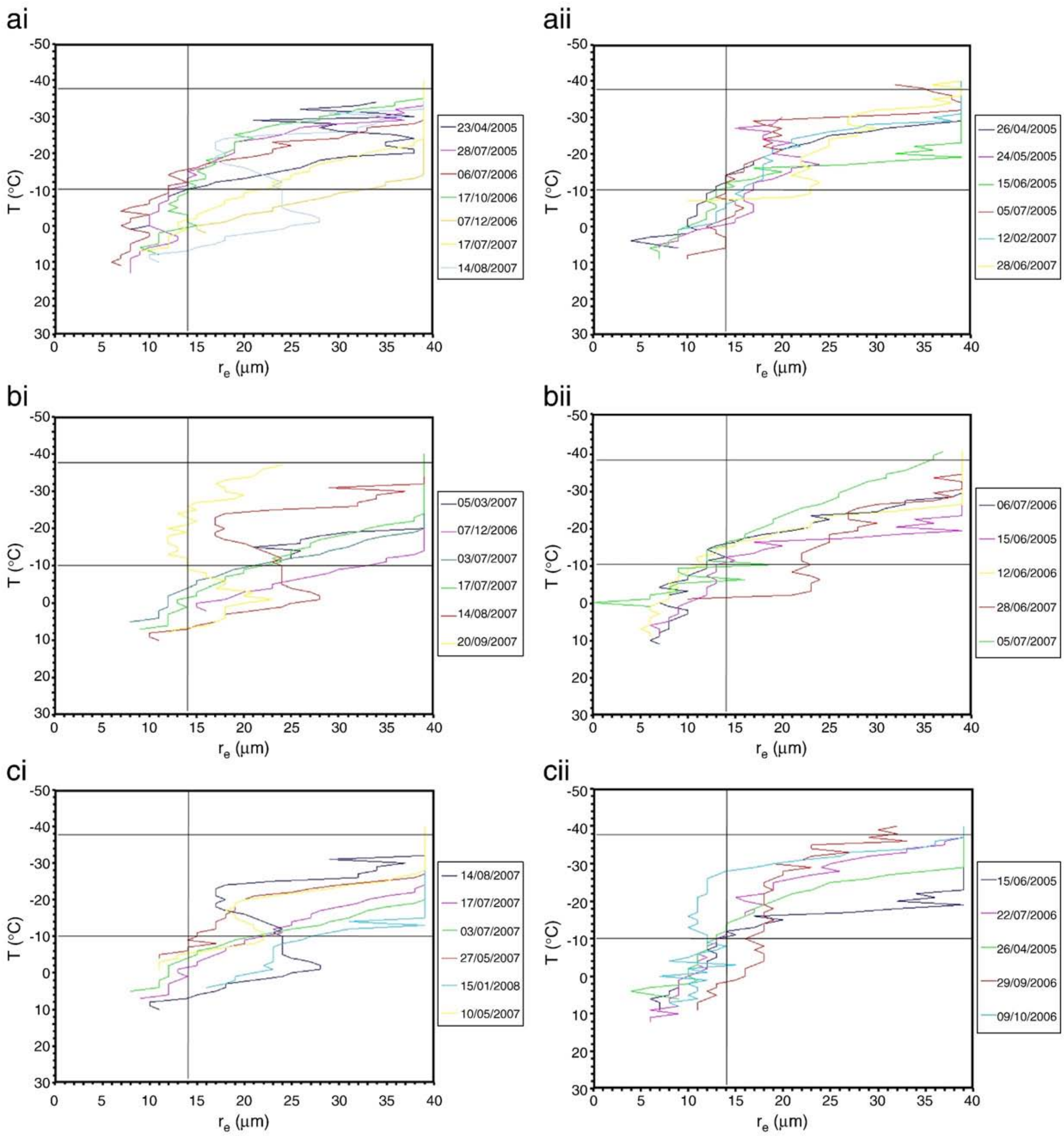

Fig. 9. T–r_e plots showing events with i) the highest fluorescence and ii) the lowest fluorescence for a) HULIS, b) TYLIS, c) TRYLIS.

Table 1
Summary of selected cases and fluorescence intensity [fluorescence intensity given as arbitrary units (a.u.)].

Event	Source	TYLIS (a.u.)	TRYLIS (a.u.)	HULIS (a.u.)
23/04/2005	Continental	342	241	471
26/04/2005	Maritime	355	125	78
24/05/2005	Maritime	398	142	85
15/06/2005	Maritime	281	70	86
05/07/2005	Maritime	384	160	79
28/07/2005	Mixed	457	363	469
12/06/2006	Continental	370	192	199
06/07/2006	Continental	380	289	481
22/07/2006	Maritime	414	121	162
29/09/2006	Maritime	381	124	238
09/10/2006	Maritime	411	140	143
17/10/2006	Continental	578	302	409
07/12/2006	Maritime	941	470	386
12/02/2007	Maritime	385	181	125
05/03/2007	Maritime	564	420	174
10/05/2007	Maritime	537	664	139
27/05/2007	Mixed	569	764	281
28/06/2007	Maritime	134	163	82
03/07/2007	Maritime	704	785	234
05/07/2007	Maritime	344	390	96
17/07/2007	Maritime	851	948	514
14/08/2007	Mixed	887	948	545
20/09/2007	Maritime	718	439	341
15/01/2008	Maritime	624	593	208

Therefore, the presence of compounds such as TYLIS and TRYLIS in microphysically 'maritime' clouds suggests that they could promote the development of ice-precipitation at 'warmer' temperatures such as in microphysically 'maritime' clouds, where large droplets freeze at relatively high temperatures. The exception to this is HULIS: a number of events containing high HULIS fluorescence are associated with both microphysically 'continental' and microphysically 'maritime' clouds — this indicates the complexity of this substance. It may be that certain types of HULIS are associated with particular types of clouds, since complex properties of organic compounds result in diverse CCN behaviours. They can introduce competing effects on the activation of droplets (Kanakidou et al., 2005). For example, film-forming compounds can inhibit the rate of condensation and evaporation and the resulting slowly-growing CCN can either decrease or increase droplet number depending upon how the material is distributed among the size distributions — this may eventually lead to a wider droplet spectra and the formation of larger droplets that initiate collision–coalescence and precipitation (Chen and Lee, 1999; Feingold and Chuang, 2002; Rudich, 2003; Broekhuizen et al., 2004; Graber and Rudich, 2006), or may result in a narrow spectra with smaller droplets. These different characteristics may give rise to either microphysically 'maritime' or 'continental' clouds. However, the lack of a specific microphysical cloud structure which is associated with low or high HULIS fluorescence values may also be reflective of below-cloud, local contributions.

Feng and Möller (2004) found the concentration of organic species to be greater in smaller cloud droplets than in larger droplets, attributed to smaller droplets originating from CCN containing more organics; smaller droplets absorbing more gaseous organics due to larger surface area–volume ratio; and/or large droplets developing from smaller droplets by condensation growth, resulting in dilution. Since the concentration of inorganic species has been found to be larger in bigger drops, they concluded that smaller drops are associated with more organic species. In some cases microphysically 'continental' clouds, with a greater number of smaller droplets, are associated with high HULIS fluorescence, which would support this theory. However, since microphysically 'maritime' clouds, characterised by larger droplets, are mainly associated with higher TYLIS and TRYLIS (and on occasions, HULIS), this appears to be the opposite of what has been found by Feng and Möller (2004). It has been discussed that TYLIS and TRYLIS, acting as ice nuclei, could promote the development of ice-precipitation at 'warm' temperatures, which may indicate that this substance is found mainly in ice crystals rather than cloud water droplets. Larger droplets also promote riming, and rimed crystals have also been found to contain higher pollution loadings compared to unrimed particles (Collett et al., 1991). Once again, the contribution from sources below the cloud cannot be ignored.

3.1.3. Source area

Certain events have a microphysically continental (maritime) structure and are, as expected, continentally- (maritime-) sourced. However, there are occasions when this is not the case. There appears to be no consistent relationship between events having a particular source area and cloud microphysical structure (Fig. 10), thus providing evidence for the influence of local areas on cloud structure. On some occasions maritime-sourced events are heavily influenced by the local geography and/or terrestrial and anthropogenic inputs, and develop a microphysically continental structure — therefore they do not have a distinctive T–r_e relationship. This was also noted by Rosenfeld and Lensky (1998) who observed transformation in air masses moving into areas affected by biomass burning smoke or urban air pollution that in these circumstances, coalescence and precipitation may be suppressed.

Continentally-sourced events however, appear to show a more distinctive T–r_e relationship, similar to conceptual 'microphysically continental' curves. Fig. 10 also shows the T–r_e plots for continentally sourced events. However, since most events travel over both maritime and continental areas, there were only a few events which spent little time over maritime areas, therefore only four true continentally-sourced events could be included. Hence, it is difficult to determine whether these events are representative of all events originating or travelling mainly over continental areas.

In order to investigate the relationships below the cloud, precipitation parameters are investigated using MRRs in the next section.

3.2. Precipitation microstructure — drop size distributions

3.2.1. Microphysical classification

The DSD curves were generated for all the events that were classified as either 'continental' or 'maritime' according to the T–r_e plots. From Fig. 11 it is evident that on average, microphysically 'continental' events contain fewer smaller droplets ($<\sim1.6$ mm) and a greater number of larger droplets, compared with microphysically 'maritime' events. Although the difference is slight, it is statistically significant ($n=46$; $t=$

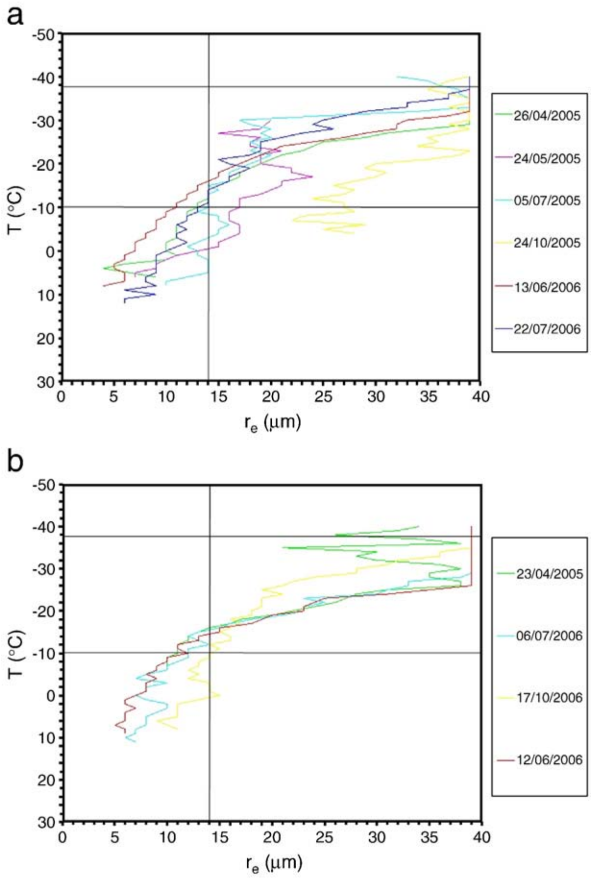

Fig. 10. T–r_e plot for a) maritime-sourced events and b) continentally-sourced events.

-2.28; $p<0.05$). When the individual plots are examined, the DSD for large number of microphysically 'maritime' events have a greater number of smaller droplets. However, there are also a number of microphysically 'maritime' events which have a DSD similar to microphysically 'continental' events. Therefore, although the average plots provide some information on the distribution of the DSD classified by microphysical structure, it is clear that there are cases which overlap.

In a rising air parcel with active coalescence as for 'maritime' clouds, the initial dominant process would be widening of the cloud drop size distribution (CDSD) into large concentrations of drizzle-sized drops, growing asymptotically to the rain drop size distribution as drizzle coalesce with drizzle, and cloud drops become raindrops, with a small median volume drop diameter, ($D_{0e}=1.76$ mm, Hu and Srivastava, 1995). During the growth phase of raindrops, R also increases with median drop diameter (D_0). R increases with fall distance from cloud top, due to growth by accretion and coalescence, until raindrops become sufficiently large enough for breakup to occur (Rosenfeld and Ulbrich, 2003). Conversely, microphysically 'continental' clouds are characterised by narrow CDSD thus having little drop coalescence and warm rain (Rosenfeld and Ulbrich, 2003). Most raindrops originate from the melting of ice hydrometeors that are typically graupel or hail in the convective elements, and snowflakes in the mature or stratiform cloud. Graupel and hail grow without breakup while falling through the supercooled portion of the cloud and grow by accretion in the warm part of the cloud, where they melt. Large melting hail stones shed the excess melt-water in the form of a DSD. Shedding stops when the melting particles approach the size of the largest stable raindrops, which are subject to further breakup due to collisions with other raindrops. New raindrop formation is therefore limited to the breakup of pre-existing larger raindrops. It is expected that in such clouds there is an excess of larger drops and a lack of small drops compared to microphysically 'maritime' clouds with active cloud drop coalescence, for a given R. These factors may also explain why precipitation associated with microphysically 'maritime' clouds has higher TYLIS fluorescence since the concentration would be less 'diluted' in the resulting smaller rain droplets.

3.2.2. Source area

Events were further categorised according to 'continental' or 'maritime' source area, as determined by back-trajectories. Fig. 12 shows the resulting mean DSDs, which have statistically significant differences ($n=46$; $t=-2.27$; $p<0.05$). On average, the continentally-sourced precipitation has fewer smaller droplets (<1.6 μm) and a greater number of larger droplets compared to maritime-sourced event. This shows a similar trend to the DSDs obtained when the events are classified according to microphysical structure.

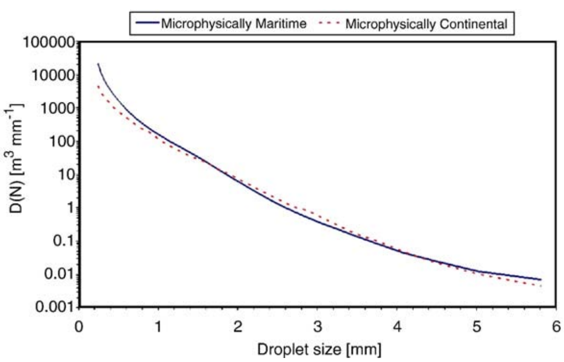

Fig. 11. Mean DSD curve for all microphysically continental and microphysically maritime events.

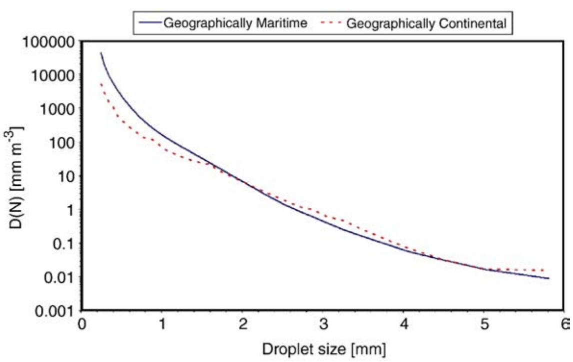

Fig. 12. Mean DSD curve for events having a geographically maritime source, compared to those having a geographically continental source area.

3.2.3. Storm type

DSD also varies with cloud dynamics: the extreme cases here being convective and stratiform. When events were classified according to their dynamic state it was found that, on average, convective events have fewer droplets <2 mm compared to stratiform events (Fig. 13). When this is broken down into individual DSDs there are a large number of convective events which have fewer smaller droplets (<1 mm) compared to stratiform events.

Rainfall in convective regions is formed as warm rain and graupel-melt, whilst stratiform precipitation is typically composed of ice particles that were advected from the convective portion of the storm, and from aggregation of newly formed ice crystals in anvils which melt into rainfall in a radar 'bright band'. DSD curves follow a similar sequence of transitions as for cloud microphysics, with convective precipitation containing a greater number of larger raindrops. One factor affecting the resulting DSD is greater updrafts where the smallest droplets are deposited in the anvil or carried aloft, especially in thunderstorms (Rosenfeld and Ulbrich, 2003). At the onset of stratiform precipitation there is an increase in D_0 due to aggregation of ice particles. Stratiform precipitation can evaporate falling from the melting layer, when small droplets evaporate preferentially.

Roy et al. (2005) suggested that the shape of the DSD curve can actually be used to help determine type of precipitation, based on the fact that riming is the main process determining the form of the DSD in convective clouds (an indication of updrafts and convection) and aggregation is the main process determining stratiform DSD. In contrast to previous work, they suggest that *for the same rainrate*, small drop DSD can be associated with convective clouds and large drop spectra with stratiform clouds (Atlas et al., 2000).

3.2.4. Fluorescence

Events were then categorised according to their fluorescence values, as in Section 3.1. Fig. 14 the mean DSD for six events with the highest fluorescence values, and six events with the lowest fluorescence values for TRYLIS. There is a significant difference ($n = 46$; $t = 2.29$; $p < 0.05$) between the mean DSD for events containing low TRYLIS and that of those containing high TRYLIS, although the differences were not significant for HULIS and TYLIS. TYLIS and TRYLIS are both amino acids, yet only TRYLIS shows a significant relationship with DSD. It is therefore likely that they may have different sources, or that they are transformed differently under certain atmospheric or droplet conditions — for example, it has been found that tryptophan is rapidly transformed in water droplets during exposure to ozone and sunlight (McGregor and Anastasio, 2001). On average, events with high TRYLIS fluorescence have a greater number of small droplets <1 mm, and fewer larger droplets. This would be expected, since microphysically 'maritime' clouds have been associated with high TRYLIS, and such clouds are associated with a DSD containing a larger number of smaller droplets. In common with other criteria, there are a number of cases which overlap when smaller droplets are considered. However, for droplets greater than approximately 1.4 mm, there appears to be a more consistent trend, whereby events containing low TRYLIS fluorescence have a greater number of larger droplets. This increase may be due to coalescence and break-up working simultaneously, a process which is more efficient in maritime clouds.

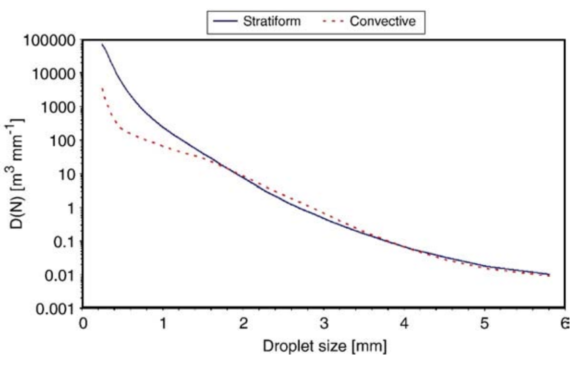

Fig. 13. Mean DSD curve for stratiform and convective events.

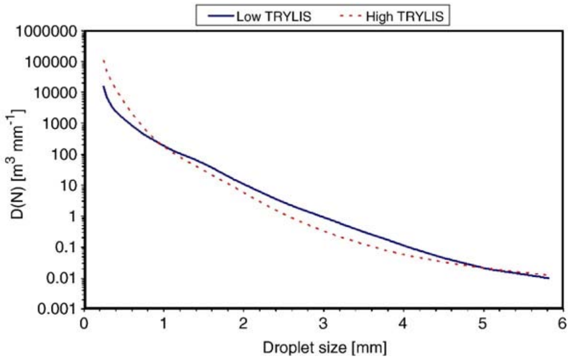

Fig. 14. Mean DSD curve for the six events containing the lowest TRYLIS fluorescence, and the six events containing the highest TRYLIS fluorescence.

The scavenging extent is also linked to aerosol concentration and size distribution, and precipitation intensity and droplet spectra. For example, low-intensity precipitation is thought to be more efficient at removing below-cloud aerosols, whilst at the same rate, precipitation with smaller droplets will remove aerosols more effectively due to larger total surface area to volume ratio (Zhang et al., 2004). Flossmann (1998) found that there was a peak scavenging efficiency at a drop radius of 0.5 mm, when particles are captured at the rear of the drop due to increasing strength of the rear drop eddy, after which the efficiency decreases again. This may also help explain why an increase in TRYLIS is observed in rainwater with a greater number of smaller droplets.

4. Conclusions and recommendations

This paper has examined a number of precipitation events using a range of complimentary techniques, and has categorized each event in order to examine the overall trends. It has described and attempted to explain the results in which CAPSAT and T–r_e plots were used to assess cloud-top microphysics, together with an array of vertically-pointing MRRs for assessing variations in DSD for categorized events. This was augmented with fluorescence spectrophotometry for examination of rainwater content. To date, T–r_e plots have been used to study deep convection, although, this study appears to demonstrate the utility of such a method in examining convective cloud structure in the mid-latitudes.

Firstly, CAPSAT and T–r_e plots were used to assess cloud microstructure for a number of contrasting events. CAPSAT alone cannot be used to distinguish the complete microphysical characteristics of events, but is a useful tool for examining various properties of storms. Events were classified as microphysically 'continental' or 'maritime' according to their T–r_e profile, by comparing these to the conceptual profiles developed by Rosenfeld and Woodley (2003). Microphysically 'maritime' events appeared to show all microphysical stages, except that of diffusional droplet growth, whilst for the microphysically 'continental' cases, small r_e (<14 μm) were observed up to and in some cases below −10 °C, indicating droplet growth was mainly through diffusional processes with no droplet coalescence zone or rainout zone identified. The point at which maximum r_e is reached indicates full development of the ice phase. For microphysically 'maritime' events, maximum r_e was reached at a relatively warm temperature (in one case −12 °C), whereas for microphysically 'continental' events maximum r_e was reached at relatively cold temperatures (between −22 °C and −38 °C), which is consistent with previous studies. The RGB composites showed how these images can be used to provide qualitative information regarding cloud composition and dynamics, but need to be used in conjunction with a quantitative technique, such as through the examination of T–r_e plots, in order to infer information of the microstructure. Two thunderstorm events, one in which a tornado touched down in Birmingham, were then examined. These had contrasting T–r_e profiles with one thunderstorm event having a more microphysically 'maritime' structure, whilst the tornadic event and the other thunderstorm had a more microphysically 'continental' structure. This was reflected in the RGB composites which indicated differences in cloud-top temperature and glaciation processes. Comparing the T–r_e plots to the conceptual model of the way in which T–r_e relationships are affected by updrafts by Rosenfeld et al. (2008) showed that these three events all had T–r_e relationships most similar to those with moderate updrafts, despite differing microphysical structure. This suggests that updrafts may be more important to the electrification of clouds than the amount of pollution or the microphysical structure of the clouds.

Events were then classified according to rainwater fluorescence and it was found that in general, high rainwater fluorescence was associated with more microphysically 'maritime clouds' — further evidence for the role of TYLIS and TRYLIS acting as ice nuclei, especially at 'warm' temperatures. In some cases however, rainwater with high HULIS fluorescence was also associated with microphysically 'continental' clouds, indicating the complexity of these substances. When source area was investigated, it appeared that continentally-sourced events had a structure similar to microphysically 'continental' clouds, yet maritime-sourced events differed in their T–r_e structure, possibly indicating the influence of local environment on their structure.

Vertically-pointing MRRs were used to examine precipitation microstructure during sampled events, particularly DSD. It appears that, even over the UK, small changes in cloud microstructure can have impacts on the resulting precipitation. Using the microphysical classification defined above, the DSD for microphysically 'continental' and 'maritime' events were compared, finding that on average, microphysically 'continental' events contain fewer smaller droplets (<1.6 mm) and a greater number of larger droplets compared to microphysically 'maritime' events. This is consistent with previous studies which have found that microphysically 'continental' clouds raindrops originate from melting ice hydrometeors or snowflakes. This melting stops when the melting particle reaches the size of the largest stable raindrop and therefore the resulting DSD has a greater number of larger droplets compared to microphysically 'maritime' clouds which have active coalescence and a large concentration of drizzle drops. When events were classified according to rainwater fluorescence, the only significant difference in DSD was found for TRYLIS, where the DSD for rainwater with low TRYLIS fluorescence had a greater number of larger droplets, possibly due to coalescence and break up working simultaneously. This is also greater for microphysically 'maritime' events. Continentally-sourced events appeared to have fewer smaller droplets and a greater number of larger droplets compared to maritime-sourced events, similar to the microphysical classification. When storm dynamics was examined, convective events appeared to contain fewer smaller droplets compared to stratiform events. In brief, continentally-sourced, microphysically 'continental', convective events with low rainwater TRYLIS, have a DSD containing fewer smaller droplets, whilst maritime-sourced, microphysically 'maritime', stratiform events with high rainwater TRYLIS have a DSD containing a greater number of smaller droplets.

From this study it is clear that there are some differences between the DSD for different events when classified according to source, cloud microphysical structure and dynamic state. However, the differences do appear to be rather small in comparison to previous studies carried out elsewhere and vary for each event. It is likely that the structure of precipitation events occurring over the UK vary to a lesser extent than events occurring over other regions. This may be due to the location of the UK where many events are 'transitional' in nature, and consequently do not show wide variations. Nevertheless, these studies are essential for improving our understanding of the variations in DSD between events and for improving Z–R law parameters. There are systematic drifts in Z–R relationships for a given rain rate: for example, R for a given Z is greater by a factor of more than 3 for maritime precipitation compared to continental precipitation, and by a factor of 1.5 to 2 for stratiform compared to convective precipitation (Rosenfeld and Ulbrich, 2003) and are primarily related to differences in DSD. The complimentary SEVIRI and MRR data are clearly useful for analyzing clouds and precipitation, in that they provide a wealth of information to allow microphysical parameters to be investigated in detail. The use of techniques for examining clouds over small areas would be improved further if complemented by simultaneous observations and data collection at cloud height, since it seems the extent to which accurate conclusions can be made from these observations alone is limited at present. Such research is essential to improve our understanding of the roles clouds play in regulating climate.

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