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Mid-tropospheric supercooled liquid water observation consistent with nucleation induced by a mountain lee wave

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[1] A case study relative to the observation of unexpected liquid water in an apparently cloudless atmosphere is presented. Microwave radiometer profiler observations on 14 April 2008 at Boulder, Colorado, USA, showed an increase in the liquid water path with values higher than 0.05 mm and corresponding relative humidity saturation from 4.75 to 6.75 km above the ground level in profiles retrieved using a neural network algorithm. The formation of small supercooled droplets identified in the microwave retrieval of the temperature and relative humidity vertical profiles may result from nucleation stimulated by a mountain lee wave. The presented analysis reveals the existence of supercooled liquid water in the mid troposphere related to a wave activity that occurred in a sky condition classifiable as "clear" and describes an atmospheric scenario consistent with the observation of the so-called twilight zone. **Citation:** Madonna, F., F. Russo, R. Ware, and G. Pappalardo (2009), Mid-tropospheric supercooled liquid water observation consistent with nucleation induced by a mountain lee wave, *Geophys. Res. Lett.*, *36*, L18802, doi:10.1029/2009GL039545.

1. Introduction

[2] From a thermodynamic standpoint, condensation of water in the atmosphere can occur under different conditions. The formation of cloud droplets in the atmosphere occurs by means of heterogeneous nucleation where aerosols act as condensation nuclei (CCN) reducing to 1–2% the supersaturation ratio required for a water vapour to liquid water phase transition. In absence of CCN, homogeneous nucleation requires very high supersaturation conditions for the development of a cloud and this probably occurs only in a laboratory environment. In the atmosphere, homogeneous nucleation has been observed for ice [*Heymsfield and Miloshevich.*, 1993; *Pruppacher*, 1995], but no experimental evidence of vapour to liquid phase transitions in absence of CCN is reported in literature.

[3] In this paper, we present an interesting case study relative to the observation of an unexpected increase in liquid water path (LWP) retrieved on the 14 April 2008 by an MP-3000A microwave profiler operating in Boulder, CO, USA. The observations showed an increase of the atmospheric liquid water content in apparently cloudless sky conditions and corresponding supersaturated conditions retrieved using a neural network profiling algorithm in a vertical region located between 4.75 and 6.75 km above the ground level (a.g.l.) with temperatures between 250 and 235 K. The lowest temperature at which liquid droplets exist for times longer than a fraction of seconds depends on the drop size [*Rosenfeld and Woodley*, 2000], but evidence of the existence of supercooled liquid water droplets down to −40°C obtained using ground based, airborne and satellite observations is reported in literature [e.g., *Sassen et al.*, 1985]. The absence of "visible" clouds has been also verified using the images of the NCAR sky webcam from the Foothills laboratory, located 1 km north of the radiometer site. The rapid and unstable event observed in Boulder could be caused by the passage of an atmospheric mountain lee wave observable both in the time series of the temperature and relative humidity profiles. The passage of the wave, in agreement with the E/NE upper level wind direction calculated by the North American Model (NAM) induced a rapid adiabatic expansion and a following compression of the atmosphere that generated strong humidity pulses with high relative humidity variations from dry to supersaturated conditions. The formation of supercooled liquid droplets in an apparent clear sky could refer to the observation of the so-called "twilight zone" [*Koren et al.*, 2007; *Pust and Shaw*, 2008], described as a region characterized by evaporating cloud fragments and enhanced aerosol with, therefore, intermediate conditions between clear and cloudy sky. This region generates a continuum of cloud optical depth evidencing the existence of cloud fragments down to very small optical depths, even lower than the aerosol background levels [*Charlson et al.*, 2007]. The presented case study shows that the passage of an atmospheric mountain lee wave in a stable atmosphere could be responsible for the occurrence of high supersaturated conditions in the free troposphere and leads to the growth of droplets in a supercooled environment. The unstable humidification and evaporation processes generated by the wave passage are consistent with the thermodynamic scenario typically observed in the twilight zone.

2. Observations and Discussion

[4] On 14 April 2008 the MP-3000A microwave radiometer profiler (E. Campos et al., Cloud water-phase dynamics observed by microwave profiling radiometry and vertically-pointing radar, submitted to *Journal of Applied Meteorology and Climatology*, 2008) operational in Boulder (105.25W, 40.02N, 1635 m a.s.l.) observed an unexpected amount of liquid water during the time period ranging from 15 to 21 UTC even though no evidence of the presence of clouds is provided by an infrared thermometer (9.6–11 μm)

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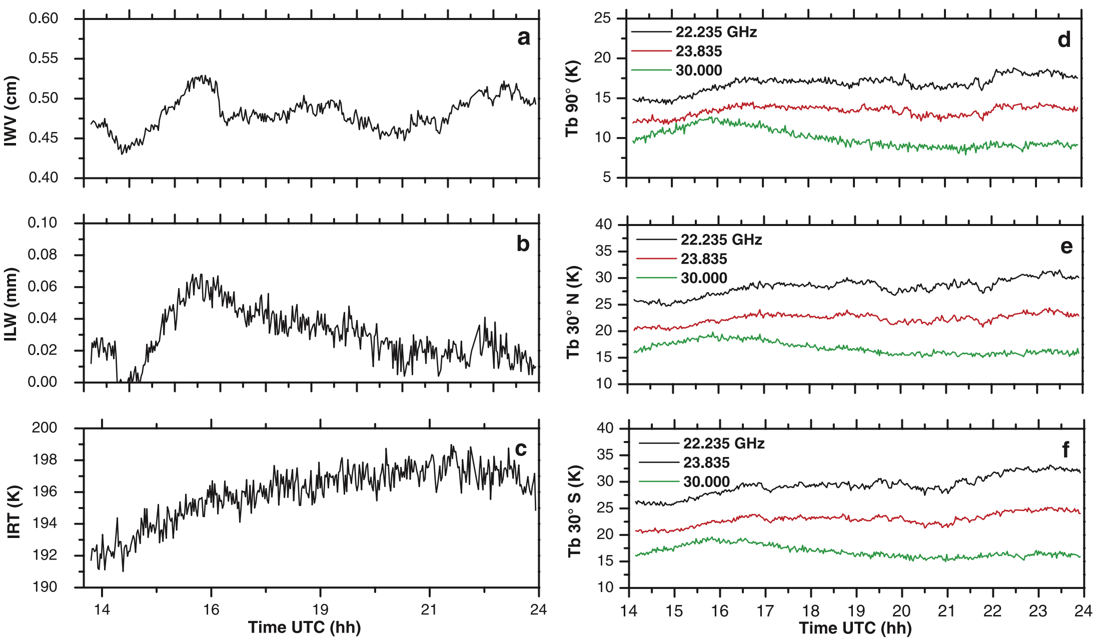

Figure 1. Time series of the (a) integrated water vapour, (b) integrated liquid water, (c) zenith infrared temperature, (d) 22.235, 23.835 and 30 GHz brightness temperatures observed at 90 deg, (e) 30 deg elevation north and (f) 30 deg elevation south measured by the MP-3000A microwave profiler on 14 April 2008 at Boulder.

included in the profiler system. From a meteorological point of view, this event is difficult to be precisely identified but we expect to have observed some kind of haze. The liquid water path (LWP) was retrieved using 27 neural net inputs including 7 upper V-band at 90 deg elevation, 7 V-band and 5 K-band data at 30 deg elevation north and south (NS), plus 58.8 GHz data at 9.45 and 19.35 deg NS, and 58.8 plus 57.288 GHz data at 41.85 deg NS. The combination of angle scanning and zenith observations can improve the upper tropospheric relative humidity retrieval since for stratified atmospheric conditions the radiometer observes twice as much integrated vapour and liquid at 30 deg elevation and it is therefore twice as sensitive to small LWP variations. The integrated water vapour (IWV) and the LWP time series from 14:07 to 23:58 UTC on 14 April 2008, and the corresponding zenith infrared temperature observation are shown in Figure 1. The LWP time series (Figure 1b) shows a significant increase after 15 UTC with peak values higher than 0.05 mm between 16:04 and 17:16 UTC.

[5] A quantification of absolute errors in microwave retrievals of cloud liquid water content is difficult. *Korolev et al.* [2007] presented a bias in the liquid-water paths retrieved from microwave radiometer observations of about 0.1 mm, obtained by comparing a 2-channel radiometer (37 and 85 GHz) with in situ (aircraft) observations. In our neural network retrievals of cloud liquid water contents, which includes scanning measurements, the values of the LWP lower than 0.02 mm are representative of clear skies and will be disregarded as retrieval noise. This LWP error estimation is similar to that obtained by *Huang et al.* [2008] using cloud tomography. However, the increase in the LWP occurring in the considered time window is significantly higher than the noise level and is representative of a real atmospheric condition.

[6] In Figure 1, the time series of the 22.234, 23.834 and 30 GHz brightness temperatures (Tb) measured by the MP-3000A at the 90 deg, 30 deg elevation north and south for the considered period are also shown. These frequencies are located in 22 GHz water vapour resonance band and, in particular, the 30 GHz frequency is the nearest of the MP-3000A to a window spectral region and therefore the most sensitive to the LWP variations.

[7] In Figure 2, the time series of temperature, water vapour mixing ratio and relative humidity profiles observed by the microwave profiler in Boulder for the same time period of Figure 1 are shown. Temperature, humidity and liquid water retrievals shown in this paper are obtained using a neural network algorithm [*Solheim et al.*, 1998] trained by forward modelling several years of historical operational radiosonde data with a radiative transfer model. The neural network algorithm retrieves temperature, vapor density, relative humidity and liquid profiles independently. Each profile is output at 58 height levels, with a vertical step of 50 m from the surface to 0.5 km in height, 100 m between 0.5 and 2 km, and 250 m between 2 and 10 km. The accuracy of the neural network retrieval applied to zenith measurements only is reported by *Ware et al.* [2003]. The root mean square (r.m.s.) deviation of the radiometer temperature profiles from the radiosoundings provided by the RAOB Denver station increases from 0.5 K to 2 K at altitudes lower then 1 km a.g.l. to about 2 K up to 5 km, while it is lower than 2.5 K above 5 km. The accuracy of the humidity profiles ranges within 0.5–1.2 g/m^3 below 4 km, where the humidity field has a strong horizontal variability, while it decreases with the height and is lower than 0.5 g/m^3 above 4 km. From the use of the elevation scanning measurements, improvements in the accuracy and vertical resolution of the retrieved profiles are expected [*Westwater et al.*, 2004]. This improvement can be limited

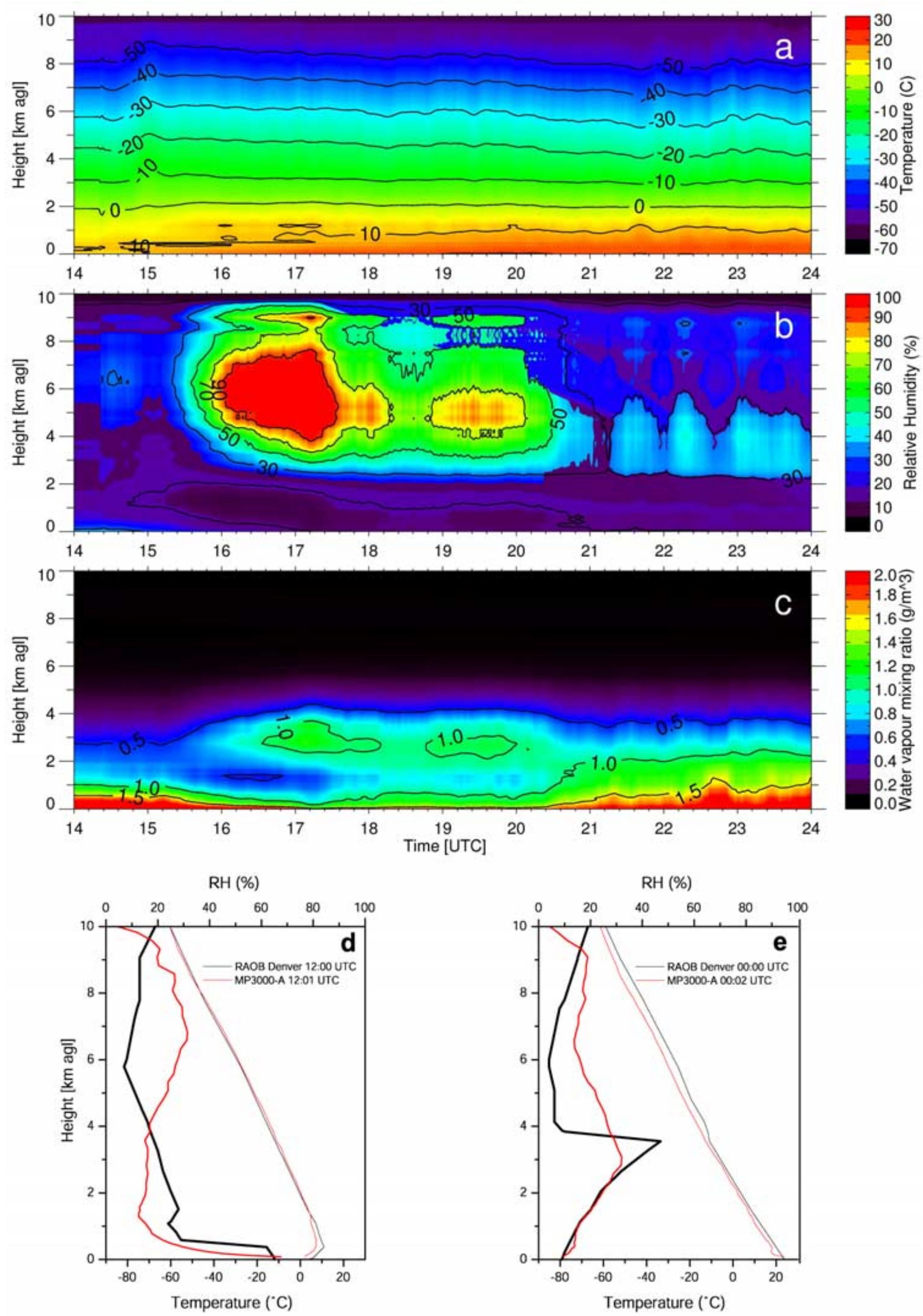

Figure 2. Time series of the vertical profiles of (a) temperature, (b) relative humidity and (c) water vapour mixing ratio up to 10 km above the ground level retrieved by the microwave profiler MP-3000A on 14 April 2008 at Boulder; comparison between the temperature (thin lines) and relative humidity (thick lines) profiles measured by the MP-3000A and the corresponding radiosounding profiles performed (d) at 00 UTC on 14 April 2008 and (e) at 00 UTC on 15 April 2008 at Denver Airport.

by different factors especially in the boundary layer. In particular, large attention in using elevation scanning measurements has to be paid for a non-linear retrieval, like the neural networks, since the horizontal inhomogeneities of the atmosphere may produce a significant bias in the retrieved humidity profiles [*Cimini et al.*, 2006]. In the presented neural retrieval, that also includes scanning measurements, additional constraints are placed on the retrieval by combining observations at appropriately selected frequencies at 30 and 90 deg elevation.

[8] One of the main issues to address in the passive retrievals is represented by the vertical resolution of the retrieved profiles, especially when investigating events that occurred at the mid- and upper tropospheric levels. In a Bayesian framework, the calculation of the degrees of freedom for a variational retrieval demonstrates how the

application of elevation scanning dramatically improves the vertical resolution of the temperature profile in the lowest 300 m but also significantly above 3 km [*Hewison*, 2006]. *Cadeddu et al.* [2002] also showed that a multi-channel system at a fixed viewing angle provides more information on the temperature profile above 1 km than a scanning single channel system. Moreover, *Cimini et al.* [2006] demonstrated how the use of scanning measurements can significantly improve humidity profile resolution in the mid-troposphere to less than 1.5 km at 5 km of altitude. The improvement of the profile vertical resolution increases the capability of retrieving vertical humidity gradients.

[9] The time series of relative humidity, reported in Figure 2b, shows the presence of humidity peaks in the upper troposphere with the occurrence of saturation conditions in a vertical region extending from about 4.75 to 6.75 km a.g.l. that corresponds to the period when the increase in the LWP is observed. The supersaturation condition observed about from 16:00 to 17:30 UTC follows a time period characterized by values of the relative humidity lower than 50% and by an increase of temperature in the considered region of about 4 K at all levels higher than 3 km a.g.l. Finally, considering the reduced profile vertical resolution in the mid-troposphere, the vertical extension of the supersaturation region estimated by the scanning retrieval, along with the low LWP value of 0.05 mm, implies a low number concentration for the observed supercooled droplets.

[10] In Figures 2d and 2e, comparisons are shown of the temperature and humidity profiles measured by the MP-3000A and the Denver radiosondes launched at 12 UTC on 14 April 2008 and at 00 UTC on 15 April 2008, 50 km SE of the radiometer location in Boulder. If we exclude the first kilometer of atmosphere, where differences at the sunrise and at the sunset over a 50 km distance and different landscapes are plausible, the agreement for the temperature profiles is very good throughout the troposphere, while there are differences in the humidity field but within the expected accuracy of the neural network retrieval. Moreover, both the Denver radiosoundings are probably affected by a dry-bias above 4 km a.g.l. [e.g., *Turner et al.*, 2003].

[11] Infrared temperature measurements (Figure 1c) by the infrared thermometer (IRT) are lower than 200 K along the whole time series, consistent with cloudless sky conditions. However, according to the literature [*Güldner and Leps*, 2005], the temperature measured by an infrared pyrometer accurately reflects the cloud base temperature if the emissivity in the spectral region near 10 microns is close to unity. This means that if we have a very low cloud fraction condition ($\ll 1$), the atmosphere is in a condition very far from the optimal working condition for the IRT, i.e., we are very far from observing a target with an emissivity equal to 1. This can cause a large underestimation of the cloud base temperature. Therefore, this means that in presence of a very small number of tiny droplets that nucleate in the supersaturated region, which is a description consistent with the presented measurement scenario, the IRT is not sensitive to the presence of liquid water in the atmosphere, though it operates in a high transparent spectral region of the atmosphere.

[12] In order to confirm the absence of clouds, images from MODIS and the NCAR webcam (available at http://www.eol.ucar.edu/webcam/img_viewer.html) have been considered. The AOD retrieved by MODIS on board of TERRA (available at http://modis-atmos.gsfc.nasa.gov) at its overpass in the vicinity of BAO at 18:35 UTC is 0.077 ± 0.003, representative of an average background value of the AOD over U.S. Moreover, the investigation of the images provided by the NCAR foothills facility fish-eye webcam, that provides one sky image per minute, clearly shows the absence of "visible" clouds during the observed supersaturation time interval. This represents a further indication of low supercooled droplet concentration in the observed supersaturation region and confirms a sky condition consistent with the detection of the twilight zone by the MP-3000A.

[13] Therefore, the question is how to explain the presence of liquid water in the atmosphere and the supersaturation conditions that occur in absence of high water vapour concentration.

[14] The microwave profiler observations reveal a scenario in agreement with the passage of an atmospheric mountain lee wave in the temperature and relative humidity profile time series. Mountain lee waves are created on the lee side of mountain ranges when strong winds hit the range at approximately a right angle. They may have horizontal wavelengths of many tens of kilometers and amplitudes of 500 m or more [*Gill*, 1982]. In correspondence of the crest of the wave, cooling by adiabatic expansion can initiate phase transition mechanisms. Mountain lee waves are often identified by the presence of interesting cloud forms, including lenticular altocumulus, forming at the crest of the wave if air reaches condensation level, but they may also form in dry air without cloud markers when the lift does not generate clouds. In our cloudless scenario, the temperature is not constant at all levels and its time series shows an increase higher than 4 K in the time window corresponding to low humidity immediately before the LWP increase (Figure 2); during the occurrence of the supersaturation conditions, the temperature slowly decays to unperturbed values. The wave passage, inducing an adiabatic expansion, generates a cooling of more than 4 K at all levels above 3 km a.g.l. Stable atmospheric conditions are observed at lower levels. Afterwards, the wave decays in a period of about 4 hours with the resulting and progressive decrease of the RH to values lower than 40% after 21 UTC.

[15] The meteorological scenario over Boulder, according to NAM data (horizontal resolution 12 km), provided by NOAA-ARL, is compatible with the formation of lee waves because it is characterized by a stable atmosphere in the downwind region while in the upwind region the wind, coming from W/SW, reaches a speed of 15 m/s at the top of the Rocky Mountains surrounding Boulder area with more than 7 m s^{-1} of difference in the wind speed at the terrain level. Moreover, model data show a wind direction perpendicular to the mountain ridge axis (within approximately 30°) up to more than 500 mbar - this is an essential condition for the development of a lee wave. These data are in agreement with the semi-empirical rules defined for the identification of strong lee waves [*Shutts*, 1997].

[16] The nature of orographically induced waves can create horizontal inhomogeneities in the considered measurement scenario, especially in the humidity field. This could increase the uncertainties in the retrieved profiles of temper-

ature and humidity since the inhomogeneities are in conflict with the usual assumption of horizontal stratification characterizing passive retrieval of atmospheric parameters. The Tb time series, reported in Figure 1, confirm the presence of inhomogeneities due to the lee wave passage since the increase of Tb at 30° elevation angle should be larger than the Tb increase observed at the zenith by a factor of 2, while the effective increase is lower. This effect is observed at all of the considered frequencies. The investigation of tipping curves provided by a co-located 12-channel microwave profiler (not shown) confirms the existence of moderate inhomogeneities in the time period ranging between 14 and 17 UTC, with a maximum effect observed in correspondence of the maximum oscillation of the temperature around 15 UTC, generated by the lee wave passage. However, the comparison of Tbs measured at N and S directions reveals a good agreement with differences lower than 0.4 K, i.e., lower than the instrumental noise level. Therefore, the retrieval is not significantly affected by inhomogeneities in the humidity field related to wave passage even though it is representative of larger atmosphere volume. This is probably due to the fact that the inhomogeneities induced by the passage of the mountain wave are, in a first approximation, symmetrically distributed along the N–S axis. This hypothesis is also supported by the wind direction provided by the NAM model. This symmetric structure of the humidity field could explain the observed increase of the Tb at the zenith and at N–S viewing directions.

[17] In summary, the formation of supercooled liquid water droplets, rapidly evaporating, has been observed by a microwave profiler in Boulder in correspondence with supersaturated conditions observed in the atmospheric region between 4.75–6.75 km. This event is generated by lee wave conditions clearly evident in both the temperature and relative humidity vertical profiles retrieved by the microwave profiler and consistent with the NAM wind analysis. The rapidity of the phase transition from supersaturated vapour to liquid and back to vapour, observed in the time series of the liquid water content measured with the microwave profiler, shows the unstable nature of the observed water phases and this behaviour is consistent with a wave-driven nucleation mechanism. The passage of the mountain lee wave induces an increase of the relative humidity up to levels that are sufficient for the growth of the supercooled droplets with the consequent occurrence of the supersaturation conditions. The subsequent decay of the lee wave progressively restores an unperturbed thermal state of the atmosphere with the consequent evaporation of the droplets. The resulting unstable equilibrium between the nucleation mechanism and the thermal modifications induced by the lee wave generates the humidity pulses observed in Figure 2. Such an observation can be best explained with the occurrence of the so-called twilight zone. The NAM model is not sensitive to this event even though its horizontal resolution is 12 km and the observed event is extended on a sufficiently large horizontal domain as demonstrated by the microwave scanning elevation measurements reported in Figure 1.

[18] Further investigations such as the development of a dedicated simulation model are required to definitely understand if a homogeneous nucleation mechanism is possible in the atmosphere and if the atmospheric conditions it requires are similar to those observed in this case study. However, the presented case study shows an interesting scenario where the adiabatic expansion and compression processes induced by a mountain lee wave passing through a stable atmosphere are able to generate supersaturated conditions and formation of small supercooled liquid droplets in low concentration. This might also indicate a probable physical connection between the mountain lee wave activities and the presence of the twilight zone. This analysis provides interesting remarks for the atmospheric modelling community considering that small amounts of supercooled liquid water are particularly interesting both for climatological and meteorological studies [*Dupont et al.*, 2008; *Knupp et al.*, 2009]. Radiative fluxes at the Earth's surface are very sensitive to small LWP changes if clouds with LWP values smaller than 50 g/m^2 are present [*Turner et al.*, 2007]. In addition, the presented case study confirms the high sensitivity of the microwave profiling to supercooled liquid water (of particular interest for modelling), the capability of the method to detect the effect of lee waves on the thermodynamic state of the atmosphere, and its sensitivity to events on small temporal scales.

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