



Satellite-based constraints on temperature-dependent aerosol–cloud feedback over a clean boreal forest and a rural continental site

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Abstract.

Temperature-dependent biogenic aerosol formation has been proposed as a negative climate feedback mechanism. However, the indirect cloud-albedo radiative feedback remains difficult to quantify from observations because in situ observations from clouds are sparse and satellite-derived cloud properties are sensitive to retrieval assumptions, cloud regime, and meteorology.

- 5 In our attempt to tackle this issue, we combine multiyear field observations with carefully sampled satellite retrievals for two diverse locations: a boreal forest site, Hyytiälä in Finland, and a continental site, Southern Great Plains in USA. We focus on low-level liquid clouds and evaluate the impact of sampling criteria (general cloud-quality filters, a local quality filter, and an adiabatic-consistency criterion) and different fitting methods on the estimation of the temperature-dependent radiative effects of biogenic aerosols. We found that if the adiabaticity requirement is not used, the indirect radiative effects are likely
- 10 underestimated because non-ideal cloud scenes dilute the observed aerosol-cloud response. Based on carefully selected data sets, we estimate that the clear-sky direct radiative feedback caused by biogenic aerosols is approximately $-0.6 \text{ Wm}^{-2}\text{C}^{-1}$ in a clean environment and about half of that ($-0.3 \text{ Wm}^{-2}\text{C}^{-1}$) in a rural region. The indirect radiative feedback of these aerosols is in the same range with the direct radiative feedback ($-0.6 \text{ Wm}^{-2}\text{C}^{-1}$) in the clean region but it is almost three times stronger ($-0.8 \text{ Wm}^{-2}\text{C}^{-1}$) in the rural region. However, the quantification of the temperature-dependent aerosol–cloud
- 15 feedback is more straightforward in clean environments than in rural ones, where other emissions can drive the aerosol load and number.

1 Introduction

Atmospheric aerosols influence the Earth’s radiation budget through both aerosol–radiation and aerosol–cloud interactions. In liquid clouds, an increase in aerosol particles that can act as cloud condensation nuclei (CCN) can increase cloud droplet

20 number concentration (CDNC), reduce droplet effective radius, and enhance cloud reflectivity, commonly referred to as the Twomey effect (Twomey, 1991). These aerosol-induced changes may also affect precipitation efficiency, cloud lifetime, and



cloud radiative properties (Albrecht, 1989). Despite decades of research, aerosol–cloud interactions remain one of the largest sources of uncertainty in the estimates of effective radiative forcing and climate sensitivity (Bellouin et al., 2020; Legg, 2021; Rosenfeld et al., 2023). A central difficulty is that aerosol effects on clouds are strongly entangled with meteorology, cloud regime, cloud dynamics, and observational uncertainty.

Natural aerosols are particularly important for constraining aerosol radiative forcing because they define the background state against which anthropogenic perturbations are evaluated (Carslaw et al., 2013). In forests and other vegetated regions, biogenic volatile organic compounds (BVOCs) emitted by vegetation can oxidize to form low-volatility vapors, contribute to secondary organic aerosol formation, and promote particle growth to CCN-relevant sizes (Hallquist et al., 2009; Kulmala et al., 2014). Since BVOC emissions and secondary aerosol formation often increase with temperature, forests may provide a temperature-dependent aerosol feedback pathway: warming can enhance aerosol loading and CCN, which may increase CDNC and cloud albedo, producing a negative radiative feedback (Paasonen et al., 2013; Mielonen et al., 2018; Yli-Juuti et al., 2021). This aerosol feedback, combined with the capacity of these ecosystems to sequester and store carbon, strengthens their potential to mitigate climate change (Kulmala et al., 2020, 2024).

Observational evidence for this pathway has strengthened in recent years. Yli-Juuti et al. (2021) showed that organic aerosol loading and CCN concentrations increase with temperature in the boreal environment and suggested that this response can produce a negative aerosol radiative feedback. However, the indirect cloud-albedo feedback remains less well constrained, because CDNC is controlled not only by aerosol number but also by updraft velocity at the cloud base, entrainment, and drizzle/precipitation. Recent in-situ evidence further suggests that low-level stratiform clouds can be highly sensitive to changes in CCN, with susceptibilities larger than many satellite-based estimates (Virtanen et al., 2025). This implies strong aerosol–cloud responses, although the local nature of in situ observations limits their applicability to the global atmosphere.

Satellite observations provide the spatial and temporal coverage needed to investigate aerosol–cloud interactions over long periods, but they also introduce important challenges. CDNC is commonly estimated from passive satellite retrievals of cloud optical thickness (COT) and cloud effective radius (CER), using assumptions about liquid-cloud vertical structure and adiabaticity (Nakajima and King, 1990; Boers et al., 2006; Quaas et al., 2006). These assumptions may not hold in heterogeneous, broken, multilayer, mixed-phase, or non-adiabatic cloud scenes. Retrieval errors in COT and CER, cloud-edge effects, viewing geometry, and natural cloud heterogeneity can propagate into CDNC and liquid water path estimates, weakening or biasing inferred aerosol–cloud relationships (Grosvenor et al., 2018; Quaas et al., 2020; Gryspeerdt et al., 2022b). Arola et al. (2022) showed that natural cloud variability and satellite retrieval errors can conceal aerosol effects on clouds, while Kokkola et al. (2025) demonstrated that spatial variability, retrieval noise, and violations of the adiabatic assumption can bias satellite-based cloud responses. These studies highlight the need for carefully constrained satellite sampling when using CDNC to quantify aerosol-cloud susceptibility.

In this study, we quantify temperature-dependent aerosol radiative feedbacks over two distinct land environments: the boreal forest site in Hyytiälä, Finland and the continental Atmospheric Radiation Measurement Southern Great Plains site in Oklahoma, United States. Building on the boreal aerosol feedback framework of Yli-Juuti et al. (2021), we use a combined satellite, in situ, and process-model approach to investigate how temperature-dependent aerosol changes affect cloud droplet

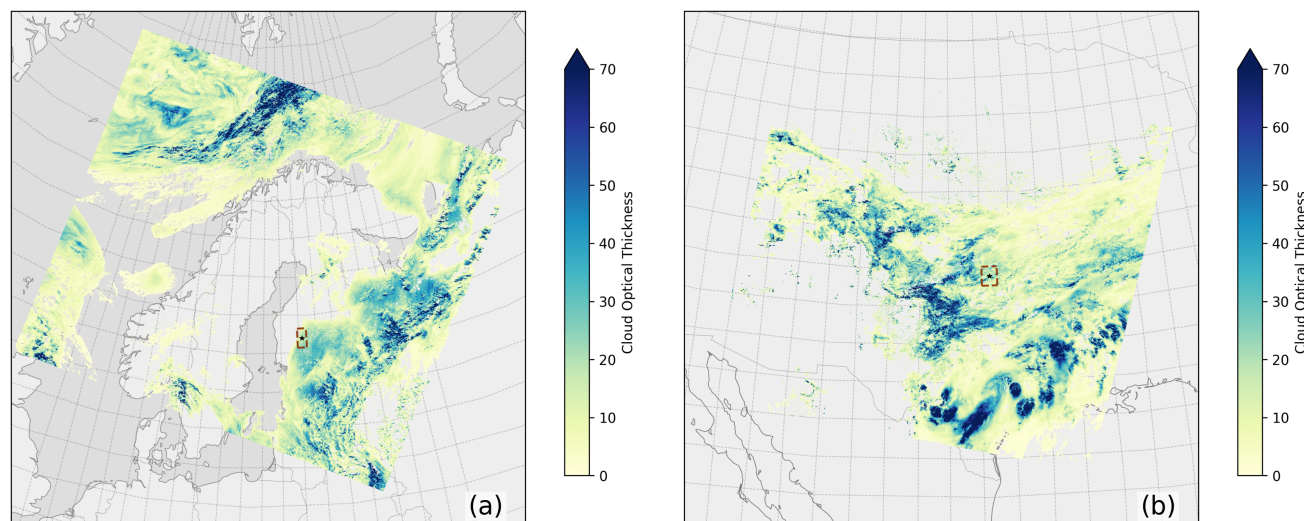


Figure 1. Terra MODIS Level-2 cloud optical thickness (COT) over (a) Hyttiälä (24-07-2013, 10:20) and (b) SGP (07-08-2017, 17:35), shown to illustrate the site locations and $1^\circ \times 1^\circ$ sampling domains. Black star marks the sites, and dashed boxes indicate the domains used for extracting MODIS cloud properties.

number and radiative feedback. Moderate Resolution Imaging Spectroradiometer (MODIS; Platnick et al., 2017) Level-2 cloud retrievals are sampled using physically constrained criteria to improve the reliability of the derived CDNC and aerosol–cloud susceptibility estimates.

60 This study focuses on two linked aerosol feedback pathways. First, we examine how temperature-dependent changes in aerosol optical properties contribute to the direct aerosol radiative feedback. Second, we investigate how temperature-dependent changes in aerosol number can influence cloud droplet number concentration and cloud albedo. The resulting framework provides a physically constrained satellite-in-situ-model estimate of direct and indirect cloud-albedo radiative feedbacks over land and evaluates how sensitive these estimates are to cloud sampling criteria.

65 2 Methodology

2.1 Sites and datasets

This study focuses on two distinct terrestrial environments to investigate how temperature-dependent aerosol changes influence cloud microphysics and radiative feedback: Hyttiälä, Finland, and Southern Great Plains (SGP), USA (shown in Fig. 1).

At Hyttiälä, long-term in situ aerosol measurements were obtained from the Station for Measuring Ecosystem–Atmosphere
 70 Relations (SMEAR II; $61^\circ 51' \text{N}$, $24^\circ 17' \text{E}$; 180 m above sea level), a boreal forest site in southern Finland. The site is located in a relatively homogeneous Scots pine stand at the Hyttiälä Forestry Field Station of the University of Helsinki and was



designed to support continuous, comprehensive measurements across the forest–atmosphere interface (Hari et al., 2012). We utilized particle number size distribution data measured with a differential mobility particle sizer (DMPS), which measures the atmospheric aerosol size distribution from 3 to 1000 nm in electrical mobility diameter. The aerosol sample was drawn from 8 m height inside the canopy, and the full size distribution was available at 10 min time resolution (Petäjä et al., 2022; Vana et al., 2016). For this study, the aerosol particle number size distributions are analyzed over a 12-summer period (2012–2023), restricted to mid-summer months (July–August). This seasonal restriction is used because boreal vegetation has a strong seasonal cycle and biogenic VOC (BVOC) concentrations (including monoterpenes and sesquiterpenes) are strongly temperature dependent and hence peak during summer (Heikkinen et al., 2023; Yli-Juuti et al., 2021; Mielonen et al., 2018). Following Yli-Juuti et al. (2021), careful separation of clean-sector air masses by exclusion of wind sector with nearby pollution source was used to support a biogenic interpretation in our study.

For SGP, the aerosol number size distribution measurements were obtained using a Scanning Mobility Particle Sizer (SMPS; TSI 3938, Minnesota, USA) at the Atmospheric Radiation Measurement (ARM) Central Facility site in north-central Oklahoma (36°38' N, 97°28' W; 315 m above sea level). The instrument measures number concentrations of aerosol particles with electrical mobility diameters in the size range of 10 to 500 nm at a temporal resolution of 5 minutes, following the methodology described by (Singh and Kuang, 2024). The summer period for this site was selected from June–August during 2017–2023, when BVOC emissions are typically strongest because of higher temperature, radiation, and leaf area indexes, making this season most suitable for examining the influence of biogenic emissions on aerosol growth and cloud microphysics (Liao et al., 2007; Mielonen et al., 2018). The datasets for both sites were accessed from their respective data repositories (SMEAR II station, 2026; Kuang et al., 2026).

Observations with total particle number concentrations below 20 cm^{-3} or above 8000 cm^{-3} were excluded, as these values deviate substantially from the long-term variability. Hourly averages were calculated from continuous 5- and 10-min observations and then used to derive parameters of a three-modal lognormal size distribution using an automatic fitting algorithm based on Hussein et al. (2005). Hourly averages were used to enable a straightforward collocation with satellite data. Only the fits with a Pearson correlation coefficient exceeding 0.99 were retained. These fits were subsequently used to estimate the number concentration of particles with dry diameter larger than 100 nm (N_{100}). The N_{100} values derived from the fitted distributions were compared with those calculated directly from the measurements, showing statistically significant linear correlation ($p < 0.05$). These values were used as a proxy for CCN-relevant aerosols, since particles of this size are expected to activate at supersaturations of approximately 0.15–0.20 % near cloud base (Mamouri and Ansmann, 2016; Choudhury and Tesche, 2023). The derived N_{100} values were used in the analysis to have exactly the same N_{100} values in the observational data set and the parcel model simulations which were based on the fitted log-normal size distributions.

Meteorological data were obtained from site-based measurements at both study locations. For Hyytiälä, air temperature at a height of 16.8 m was measured with a Pt100 inside a ventilated custom-made radiation shield (SMEAR II station, 2026). For SGP, air temperature was obtained from the ARM Surface Meteorology Systems (MET) from the Extended Facility E13, Lamont, Oklahoma (Kyröuac et al., 2021). This facility is co-located with the SGP Central Facility, where the aerosol size distribution measurements were made, and is approximately 300 m away; therefore, the meteorological and aerosol observations



were treated as spatially representative of the same site. The ARM MET system uses conventional in situ sensors to provide 1 min measurements of surface meteorological variables, including air temperature, relative humidity, pressure, wind speed, wind direction, and precipitation.

110 2.2 MODIS cloud retrievals and sampling strategy

Cloud optical and microphysical properties were obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS, Collection 6.1 Level-2 products) onboard the Terra and Aqua satellites (Platnick et al., 2017). MODIS Level-2 retrievals were used rather than gridded Level-3 products in order to apply pixel-level sampling criteria and to retain control over the cloud scenes used in the CDNC calculation. The CDNC retrieval for warm liquid clouds relies on their representation as horizontally
 115 homogeneous cloud fields that follow the pseudo-adiabatic cloud model with vertical profiles of liquid water content that correspond to a constant fraction of the adiabatic value with vertically constant cloud droplet number concentration and droplet size distribution breadth (Nakajima and King, 1990; Boers et al., 2006; Quaas et al., 2006; Grosvenor et al., 2018; Gryspeerdt et al., 2022b). The same sampling framework was applied at both Hyytiälä and SGP, with site-specific analysis periods. For each overpass, retrievals were sampled within a fixed geographic box centered on the station location. In the main analysis, a $1^\circ \times 1^\circ$
 120 domain was used, although sensitivity tests were also performed for $0.5^\circ \times 0.5^\circ$ and $2^\circ \times 2^\circ$ domains (see the supplementary information).

Since CDNC is not provided directly in the MODIS Level-2 cloud product, it was derived from cloud optical thickness (COT) and cloud effective radius (CER) following the adiabatic formulation

$$CDNC = \gamma f(T) COT^{0.5} CER^{-2.5} \quad (1)$$

125 where $\gamma = 1.37 \times 10^{-5} \text{ m}^{-2}$ (Quaas et al. (2006) and $f(T) = 0.0192T - 4.293$ (with a temperature in K), which is the temperature-dependent condensation rate (Bennartz, 2007; Gryspeerdt et al., 2016).

To reduce retrieval artifacts and improve the physical consistency of the cloud sample, three main sampling levels were defined in this study. First, a set of general cloud filters was applied to retain pixels suitable for passive-satellite CDNC retrieval. These filters require solar zenith angle $< 65^\circ$, sensor zenith angle $< 55^\circ$, single-layer clouds, liquid-cloud phase, cloud mask
 130 spatial variability index < 30 , $4 \leq COT \leq 70$, $4 \leq CER \leq 30 \mu\text{m}$, 5km cloud fraction ≥ 0.9 , cloud-top temperature ≥ 268 K, and $50 \leq LWP \leq 300 \text{ gm}^{-2}$. The COT and CER limits exclude optically thin or extreme retrievals for which the CDNC derivation is less reliable, while the high cloud-fraction threshold reduces cloud-edge and partial-cloud contamination. The constraints in LWP reduce the influence of very thin clouds, precipitating or drizzle-affected clouds, and scenes with strongly varying cloud water. Restrictions on solar and sensor zenith angles reduce viewing-geometry-related biases in COT and CER
 135 retrievals. Similar constraints have been recommended or used in previous satellite-based CDNC and aerosol-cloud interaction studies to improve retrieval reliability (Platnick et al., 2017; Grosvenor et al., 2018; Gryspeerdt et al., 2022a). However, these criteria are applied primarily at the pixel level and do not fully account for the quality of the surrounding cloud field. A pixel may pass the general filters listed above, but still be located close to ice-cloud pixels, failed liquid-cloud retrievals, or strongly



heterogeneous cloud edges, where three-dimensional radiative effects and phase contamination can influence the retrievals. To reduce this source of contamination, we applied a second screening step, hereafter referred to as the local quality filter that removes pixels neighbouring unsuccessful liquid-cloud retrievals or ice-cloud pixels, identified from the MODIS 1-km quality-assurance information. This neighbourhood-based screening provides an additional spatial-consistency check and restricts the analysis to pixels existing within more coherent liquid-cloud regions. To our knowledge, this type of local quality filter has not been applied in previous MODIS-based CDNC–aerosol susceptibility studies. The need for such filtering is supported by recent evidence that cloud heterogeneity, retrieval errors, and non-ideal cloud structure can bias aerosol effects inferred from satellite observations (Arola et al., 2022; Kokkola et al., 2025). Finally, a third filter called the adiabatic assumption was applied to the general and local filtered sample. In an ideal adiabatic liquid cloud, droplet effective radius is expected to increase with height because liquid water content increases upward. Since the MODIS shortwave-infrared channels have different photon penetration depths, this vertical structure is commonly expected to produce wavelength dependent retrievals of CER so that $CER_{3.7\mu m} > CER_{2.1\mu m} > CER_{1.6\mu m}$, with the $3.7\mu m$ channel being weighted closest to cloud top and the $1.6\mu m$ channel sampling deeper into the cloud (Zhang and Platnick, 2011; Platnick et al., 2017; Grosvenor et al., 2018). In this study, we used the relaxed criterion of $CER_{3.7\mu m} \geq CER_{2.1\mu m}$, rather than requiring the full three-channel ordering, because $1.6\mu m$ retrieval is hindered by surface contribution, cloud heterogeneity, and retrieval noise, particularly over land. Therefore, the relaxed condition retains the main physical requirement of increasing droplet size toward cloud top while avoiding excessive sensitivity to the more uncertain $1.6\mu m$ retrieval (Rausch et al., 2017).

The final filtered dataset is therefore restricted to high-cloud-fraction, single-layer, liquid clouds with physically plausible optical and microphysical properties, reduced local retrieval contamination, and an adiabatic effective-radius structure. This sampling strategy reduces the number of available pixels and overpasses, but it improves the physical interpretability of the CDNC–aerosol relationships as it increases the likelihood that the observed clouds follow the cloud structure assumptions used in the retrieval of CER and COT.

After applying the MODIS sampling criteria, CDNC was calculated for each retained pixel using the filtered COT and CER retrievals. Terra and Aqua overpasses were combined, sorted by overpass time, and restricted to the summer months used in the analysis. For each overpass, all valid CDNC pixels within the sampling domain sizes were averaged to obtain an overpass-mean CDNC. To avoid unstable overpass statistics caused by sparse sampling, only overpasses with at least 30 valid MODIS pixels within the sampling domain were retained for the subsequent aerosol-cloud analysis.

2.3 Uncertainty estimation and fitting methods

Two complementary uncertainty components were considered for the MODIS-derived CDNC and the field observations of N_{100} : the measurement uncertainty ϵ_{meas} and the spatial or temporal variability within the sampled domain ϵ_{var} . Based on them, the total relative uncertainty was calculated as

$$\epsilon_{\text{total}} = \sqrt{\epsilon_{\text{meas}}^2 + \epsilon_{\text{var}}^2}. \quad (2)$$



The MODIS retrieval uncertainty was estimated by propagating the relative uncertainties reported by MODIS for COT and CER. Denoting the relative uncertainties in COT and CER by $\Delta COT/COT$ and $\Delta CER/CER$, respectively, the relative uncertainty in CDNC was computed as

$$\epsilon_{sat} = \sqrt{\left(0.5 \frac{\Delta COT}{COT}\right)^2 + \left(2.5 \frac{\Delta CER}{CER}\right)^2}. \quad (3)$$

175 For CDNC, $\epsilon_{meas} = \epsilon_{sat}$, where ϵ_{sat} is given by Eq. (3), and ϵ_{var} is the relative spatial variability within the overpass. The overpass-mean MODIS dataset was then collocated with in-situ aerosol size distribution and meteorological observations. For each MODIS overpass time, an exact temporal match was used when available; otherwise, in-situ observations within a ± 2 h window were averaged.

The uncertainty in the collocated in-situ aerosol quantity N_{100} was estimated separately for each MODIS overpass. For N_{100} ,
 180 ϵ_{meas} was taken as 15 % of the collocated N_{100} value. The temporal variability term, ϵ_{var} was estimated from the hourly N_{100} values within the same ± 2 h window used for the collocation. When more than one in-situ measurement was available in this window, the standard deviation of N_{100} in the window was calculated and combined in quadrature with the 15 % instrumental uncertainty. Since, hourly averaged data were used, this term may give a lower estimate of the true short-term variability in N_{100} . These uncertainty estimates in CDNC and N_{100} were used in the error-based regression analysis of the aerosol-cloud
 185 relationship.

We applied three linear regression techniques to quantify the susceptibility of CDNC to perturbations in CCN-relevant aerosol concentrations. The primary relationship of interest was the dependence of CDNC on N_{100} in logarithmic space. Ordinary least squares (OLS) regression was used as a baseline method because of its simplicity and widespread use in aerosol-cloud studies (Yue et al., 2013; Sreehari and Ghantasala, 2019). However, OLS assumes that the predictor variable is error-free
 190 and may therefore produce biased estimates when both aerosol and cloud quantities are uncertain.

To account for measurement uncertainties in both variables, we used errors-in-variables bivariate approaches, namely Deming regression (DR) and York regression (YR). In DR, both the dependent and independent variables are assumed to contain errors, and the regression minimizes the orthogonal distance to the fitted line weighted by the relative error variances (Francq and Govaerts, 2014). The DR assumes homoscedastic errors and uses a constant error-variance ratio, whereas the YR allows
 195 heteroscedastic errors by incorporating observation-specific uncertainty estimates for both variables and solving iteratively for the slope and intercept (York et al., 2004; Pitkänen et al., 2016). In the fitting procedure, the relative uncertainty estimates for CDNC and N_{100} were used in the bivariate regressions, while OLS was retained as a reference fit. The regression fitting criteria and notation are summarized in Table S1.

2.4 Cloud parcel model simulations

200 To investigate cloud droplet formation under the observed aerosol conditions, a cloud parcel model (CPM) was used to simulate the evolution of the physicochemical properties of an aerosol population during adiabatic ascent (Kokkola et al., 2003; Romakkaniemi et al., 2009). The model is a moving sectional aerosol model in which the sections are defined using a constant



volume ratio between adjacent bins. It solves the ordinary differential equations describing condensational growth of water vapour on aerosol particles together with the evolving microphysics and thermodynamics of the rising air parcel (Kokkola et al., 2006). For each simulation, the measured aerosol size distribution was represented by three lognormal aerosol modes characterized by their modal number concentrations, geometric mean diameters, and geometric standard deviations. The observed aerosol size distributions were used as model input, while the environmental conditions were held fixed throughout the simulations.

For Hyytiälä, the aerosol size distributions used to initialize the model were obtained from the DMPS measurements at SMEAR II. The aerosol population was assumed to be internally mixed, with the same bulk chemical composition assigned to all size modes. This composition was based on summertime aerosol chemical characteristics observed at Hyytiälä with aerosol mass spectrometer (AMS) measurements (Finessi et al., 2012; Corrigan et al., 2013; Riva et al., 2019; Heikkinen et al., 2020). The model aerosol composition was chosen to represent the enhanced contribution of biogenic organic compounds during summer, with organic material accounting for 66.6 % of the total aerosol mass fraction and ammonium sulfate for 33.3 % (Hao et al., 2018; Heikkinen et al., 2020), consistent with the strong influence of boreal biogenic emissions (Ivan et al., 2013).

For the SGP simulations, aerosol chemical composition was prescribed using summertime aerosol mass spectrometer observations from the same site (Parworth et al., 2015). Previous studies have shown that summertime submicron aerosol mass at SGP is dominated by organic aerosol, with sulfate as the largest inorganic component and nitrate contributing only a minor fraction. Accordingly, the aerosol composition was represented by organic aerosol, sulfate, nitrate, and ammonium, with relative mass fractions based on the observed mean aerosol mass concentrations reported by (Liu et al., 2021). In the model input, these corresponded to 66 % organic aerosol, 23 % sulfate, 2 % nitrate, and 9 % ammonium. For both sites, the same aerosol composition was prescribed for all three aerosol modes, and the specified mass fractions were normalized internally within each mode by the model. In all simulations, the aerosol population was represented using 300 size bins distributed across three lognormal size modes, allowing the model to resolve condensational growth and activation kinetics.

The model was initialized with a temperature of 277 K, pressure of 900 hPa, relative humidity of 95 %, and cloud-base height of 1000 m. These values were chosen to represent lower-tropospheric warm cloud conditions at the study sites, allowing the simulations to focus on the aerosol activation under a consistent thermodynamic setup. An updraft velocity of 0.3 m s^{-1} was set for both Hyytiälä and SGP, based on characteristic updraft velocities derived from Doppler lidar observations at the respective sites. For Hyytiälä, cloud-base updraft velocities were derived by combining ACTRIS Cloudnet target classification data with Halo Doppler lidar mean Doppler velocity measurements, processed following the methodology of Manninen et al. (2018). Liquid-containing cloud layers were identified from the Cloudnet target classification using cloud classes 1–3, and the lowest liquid layer was selected as the cloud base. Only cloud-base heights between 300 and 3000 m were retained. Doppler lidar mean Doppler velocity was then extracted from the range gate immediately below the identified cloud base. Positive vertical velocities were treated as updrafts and aggregated to hourly statistics. The resulting hourly updraft distribution was fitted with a lognormal function to characterize typical cloud-base updraft conditions over Hyytiälä. A lognormal fit was used because only positive updrafts were considered and the distribution was right-skewed.



For SGP, cloud-base vertical velocity statistics were obtained from the ARM Doppler Lidar Vertical Velocity Statistics value-added product (DLPROF-WSTATS; Newsom, 2019). The standard deviation of vertical velocity at cloud base was calculated as $\sigma_w = \sqrt{w_{\text{variance}}}$. To retain representative cloud-base turbulent conditions, the data were filtered to include only cases with cloud-base height between 300 and 3000 m, cloud-base updraft fraction greater than 0.6, and Doppler-lidar cloud frequency greater than 0.7. Hourly median values of σ_w were then calculated and used to characterize the updraft distribution. Lognormal fits to the Hyytiälä and SGP distributions indicated that $w = 0.3 \text{ ms}^{-1}$ was representative of typical cloud-base updraft conditions at both sites (see Supplementary Figure S3) thus, this value was used in all Cloud parcel model simulations.

The model was run for each observed aerosol size distribution that had concurrent cloud observations using the prescribed site-specific aerosol composition and fixed environmental initialization parameters. Activated CDNC was calculated as the number of particles with radius larger than 2 micrometers. With thermodynamic conditions and updraft velocity held fixed, the CPM simulations were used to isolate the influence of aerosol number concentration and size distribution on droplet activation and to interpret the observed CDNC–aerosol relationships at Hyytiälä and SGP.

2.5 Aerosol–temperature response and direct radiative feedback

The direct aerosol radiative feedback was estimated from the temperature dependence of aerosol optical depth (AOD), derived from AERONET observations from both sites. AERONET Cimel sun photometers provide cloud-screened spectral aerosol optical depth measurements, and have been widely used for evaluating aerosol radiative effects (Holben et al., 1998). We used Version 3, Level 2 AERONET AOD at 500 nm together with the 440–870 nm Ångström exponent. Following the screening approach used by Yli-Juuti et al. (2021), only observations with Ångström exponent larger than 0.75 were retained in order to reduce possible cloud-contaminated cases. Daily mean AOD values were calculated only for days with sufficient daytime AERONET sampling, when at least six valid AERONET observations were available during the day. Daily mean temperature was calculated from the sites’ meteorological observations and collocated with the daily AERONET AOD values. The AOD–temperature response was then estimated using multivariate regression following the approach of Mielonen et al. (2018). Based on our earlier studies (Mielonen et al., 2018; Yli-Juuti et al., 2021), the main sources of summertime aerosols in these regions are biogenic and anthropogenic emissions. Therefore, we used tropospheric NO_2 product (OMNO2d) from the Ozone Monitoring Instrument (OMI; Bucsela et al., 2013) as a proxy for anthropogenic influence. The data are available on a $0.25^\circ \times 0.25^\circ$ grid, and thus we re-gridded them into a $1^\circ \times 1^\circ$ resolution to match the MODIS cloud data. The multivariate regressions were calculated with the following equation:

$$AOD = a \text{NO}_2 + bT_c + c, \quad (4)$$

where NO_2 is the tropospheric NO_2 concentration in molecules cm^{-2} and T_c is temperature in Celsius.

Hyytiälä is a very clean location with an average daily tropospheric NO_2 concentration of $1 \times 10^{15} \text{ molec cm}^{-2}$ during the analyzed summers (2017–2023), thus the influence of anthropogenic emissions is negligible and the uncertainty in the OMI



retrievals hinders this approach. Therefore, we set the constant a to zero for Hyytiälä. The Equation (4) is also used in the estimation of the temperature dependence of N_{100} .

270 The temperature-dependent direct aerosol radiative feedback was calculated as

$$F_{\text{dir}} = S_{\text{rad}} \phi \frac{\partial AOD}{\partial T} (1 - C_c) T_{\text{atm}}^{af} (1 - A_s)^2 \left[\frac{2A_s(1 - \omega)}{(1 - A_s)^2} - \beta\omega \right]. \quad (5)$$

where F_{dir} is the direct aerosol radiative feedback in $\text{W m}^{-2} \text{ } ^\circ\text{C}^{-1}$, S_{rad} is the daily mean incoming solar radiation at the top of the atmosphere, ϕ is the mean daytime secant of the solar zenith angle, C_c is the fractional cloud amount, T_{atm}^{af} is the aerosol-free atmospheric transmission, A_s is the shortwave surface reflectance, ω is the aerosol single-scattering albedo, and β is the upscatter fraction. For the clear-sky estimate, $C_c=0$, whereas the all-sky estimate, C_c was derived from MODIS Level-2 cloud observations within each sampling domain around each site. The MODIS 1-km cloud mask was used to identify confidently cloudy and confidently clear pixels, and the cloud phase product was used to identify water-phase cloudy pixels. The water-phase cloud fraction was then calculated from the number of confidently cloudy water-phase pixels relative to the total number of confidently cloudy water-phase and confidently clear pixels.

280 For Hyytiälä, the radiative parameters were selected following Yli-Juuti et al. (2021): $S_{\text{rad}} = 398 \text{ W m}^{-2}$, $\phi=1.82$, $T_{\text{atm}}^{af}=0.76$, $A_s=0.12$, $\omega=0.92$, and $\beta=0.29$. For the all-sky calculation we used MODIS water-phase cloud fraction for the Hyytiälä domain, $C_c=0.51$.

For SGP, the same formulation was applied using radiative parameters consistent with the regional United States calculation of Mielonen et al. (2018): $S_{\text{rad}} = 461 \text{ W m}^{-2}$, $\phi=1.33$, $T_{\text{atm}}^{af}=0.76$, $A_s=0.15$, $\omega=0.972$, and $\beta=0.21$. The all-sky calculation 285 used the MODIS water-phase cloud fraction for the SGP domain, $C_c=0.49$.

The lower and upper confidence limits of $\partial AOD/\partial T$ were propagated through the radiative-efficiency equation to obtain the 95 % confidence interval of F_{dir} . Negative values indicate a cooling direct aerosol feedback, in which temperature-associated increases in AOD enhance shortwave scattering and reduce the amount of solar radiation reaching the surface.

2.6 Indirect radiative feedback

290 The indirect cloud-albedo radiative feedback was estimated by combining the aerosol-cloud susceptibility with the temperature dependence of N_{100} . This approach follows the radiative framework used in previous aerosol feedback studies, where aerosol-driven changes in CDNC modify COT and cloud albedo (Charlson et al., 1992; Yli-Juuti et al., 2021). The MODIS water-phase cloud fraction used in this calculation were derived using the same cloud-mask and cloud-phase procedure described in Sect. 2.5.

295 Cloud albedo, A_c , was estimated from the MODIS COT observations using a two-stream approximation following Bohren (1987):

$$A_c = \frac{\frac{(1-g)}{\cos\theta} COT}{2 + \frac{(1-g)}{\cos\theta} COT} \quad (6)$$



where g is the asymmetry parameter and θ is the mean solar zenith angle. The mean solar zenith angle was calculated separately for each site and season. The cloud asymmetry factor (g) was set to 0.85. A correction factor (f_s) for multiple reflections between the surface and cloud was applied as

$$f_s = \frac{(1 - A_s)^2}{(1 - A_s A_c)^2}, \quad (7)$$

and the temperature-dependent cloud-albedo change was then estimated as

$$\frac{\partial A_c}{\partial T} = f_s A_c (1 - A_c) \frac{1}{3} \frac{\partial \ln(CDNC)}{\partial \ln(N_{100})} \frac{\partial \ln(N_{100})}{\partial T}. \quad (8)$$

Here, $\partial \ln(CDNC)/\partial \ln(N_{100})$ is the aerosol–cloud susceptibility derived from the collocated MODIS CDNC and in-situ N_{100} relationship, and $\partial \ln(N_{100})/\partial T$ is the temperature dependence of aerosol number concentration. These calculations were performed using the OLS based susceptibility estimates, and separately for the adiabatic and non-adiabatic cloud samples. The corresponding indirect radiative feedback was then calculated as

$$F_{in} = -S_{rad} T_{atm}^{af}{}^2 C_c \left\langle \frac{\partial A_c}{\partial T} \right\rangle, \quad (9)$$

Negative values indicate a cooling feedback caused by temperature-dependent aerosol increases enhancing CDNC and cloud albedo. Uncertainty ranges were estimated by propagating the confidence intervals of the two fitted sensitivity terms: the CDNC– N_{100} susceptibility and the N_{100} –temperature slope.

3 Results and Discussion

3.1 Aerosol-Temperature response

To study the temperature dependence of aerosol loads at these sites in detail, we analyzed how AOD and N_{100} depend on temperature at Hyytiälä and SGP. Figure 2 shows the temperature dependence of AOD at the sites and Figure 3 shows the temperature dependence of N_{100} . The AOD–temperature relationship is used for the estimation of the direct aerosol radiative feedback, while the N_{100} –temperature relationship is combined with aerosol–cloud susceptibility to estimate the indirect cloud-albedo feedback.

At Hyytiälä, AOD increases with temperature by approximately 0.007 [95 % confidence interval, CI: 0.005, 0.009] °C^{−1}. The positive AOD-temperature response is consistent with increased biogenic emissions and secondary organic aerosol formation under warmer conditions, which enhances aerosol scattering and contributes to negative direct radiative feedback. The fitted coefficients in the Equation (4) for SGP were $a = (2.68 \pm 0.73) \times 10^{-17}$ cm² molec^{−1}, $b = 0.0056 \pm 0.0005$ °C^{−1}, and $c = (1.99 \pm 0.17) \times 10^{-4}$, indicating an AOD-temperature dependence of 0.006 °C^{−1}. As part of the AOD–temperature relationship is masked by variability in other aerosol sources, the multivariate coefficient should therefore be interpreted as a NO₂-adjusted AOD–temperature sensitivity rather than as the total observed AOD response to temperature.

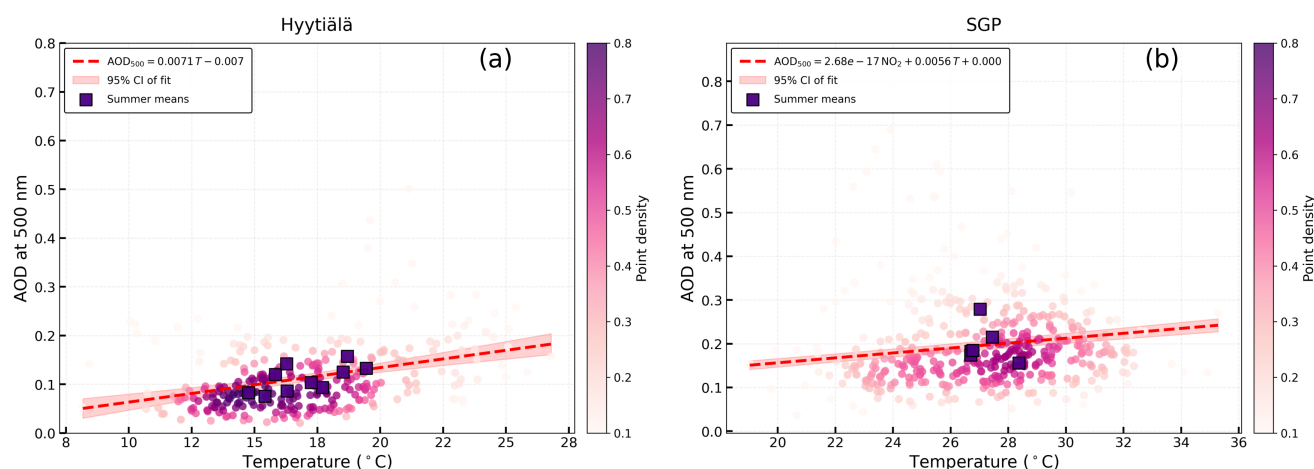


Figure 2. AOD–temperature relationships at (a) Hyytiälä and (b) SGP during summer. Point density shows the distribution of individual observations, square markers indicate summer mean values, and dashed lines show linear regression fits with 95 % confidence intervals. For SGP the fit line shows predictions from the multivariate regression as temperature varies, while the tropospheric NO₂ concentration is held at its mean value. The slopes represent the temperature dependence of AOD used in the direct aerosol radiative feedback calculation.

The temperature dependence of N₁₀₀ was positive at both sites as shown in Figure 3. At Hyytiälä, the regression analysis of ln(N₁₀₀) against temperature produced a slope of 0.138 [CI : 0.126, 0.151] °C⁻¹. At SGP, the NO₂-adjusted temperature slope was 0.241 ± 0.004 °C⁻¹. This indicates that, after separating the linear contribution associated with NO₂, temperature retains a strong positive association with N₁₀₀ also at SGP. At both sites, the linear fits match quite well with the summer means (shown with square markers in Figure 3) which could indicate that N₁₀₀ concentrations react to climatological changes in summertime temperature at these locations.

The aerosol–temperature relationships differ clearly between the two sites. At Hyytiälä, both AOD and ln(N₁₀₀) increase with temperature, with slopes of about 0.007 °C⁻¹ and 0.14 cm⁻³ °C⁻¹, respectively. These values are comparable to the ones published by Yli-Juuti et al. (2021) for the same site, but with a shorter time range. This confirms the dominant role of biogenic aerosols in this region and their potential to enhance AOD and aerosol number as temperatures increase. At SGP, the NO₂-adjusted AOD–temperature slope is 0.006 °C⁻¹ and the NO₂-adjusted ln(N₁₀₀)–temperature slope 0.24 cm⁻³ °C⁻¹, which are in the same range as the slopes in Hyytiälä. Furthermore, Mielonen et al. (2018) reported an AOD–temperature slope of 0.007 °C⁻¹ for southeastern USA, which is well in line with our estimate.

To understand why the temperature dependence of AOD is similar at both sites, but the temperature dependence of N₁₀₀ in Hyytiälä is weaker than in SGP, we studied how the aerosol size distributions differ in cold and warm temperatures at these sites (see Supplementary Figure S1). In Hyytiälä, the number of particles does not change much between cold and warm cases, but in warm cases the median size of the aerosols is significantly larger than in the cold cases, which introduces a clear AOD

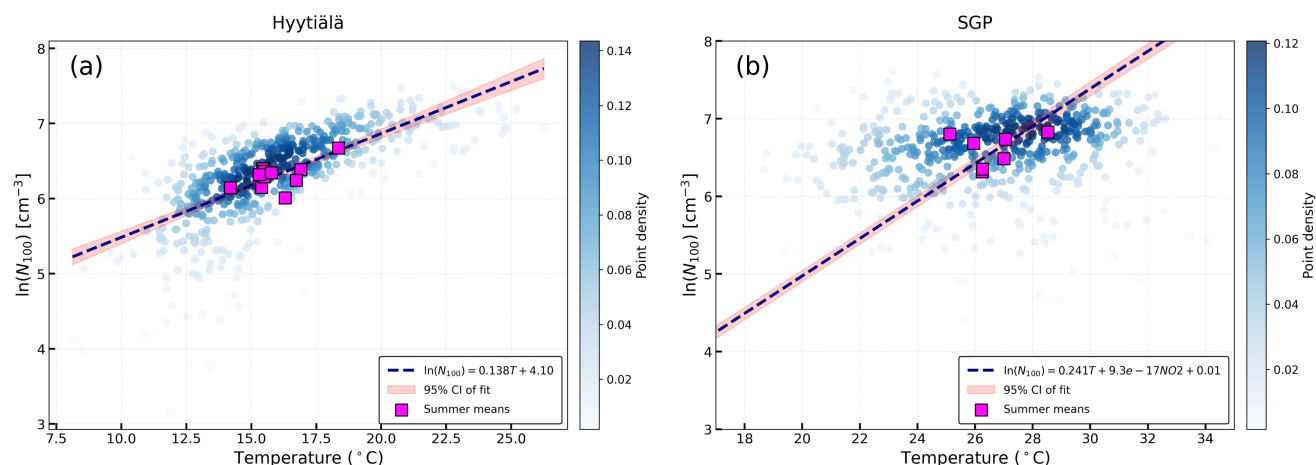


Figure 3. N_{100} -temperature relationships at (a) Hyytiälä and (b) SGP during summer. Point density shows the distribution of individual observations, square markers indicate summer mean values, and dashed lines show linear regression fits with shaded 95 % confidence intervals. For SGP the fit line shows predictions from the multivariate regression as temperature varies, while the tropospheric NO_2 concentration is held at its mean value. The fitted slopes quantify the temperature dependence of N_{100} , which is used together with aerosol–cloud susceptibility to estimate the indirect cloud-albedo feedback.

signal. At SGP, the shape of the size distributions stays similar at different temperatures, but the warm cases have significantly larger number of aerosols. Consequently, N_{100} is more sensitive to changes in temperature than AOD at SGP.

3.2 Effect of MODIS cloud sampling on CDNC retrievals

To illustrate the effect of different sampling criteria on CDNC retrievals from the MODIS Level-2 data, Figures 4 and 5 show example overpasses over Hyytiälä and SGP. The first panels show unfiltered CDNC retrievals after removal of fill values, and therefore represents the maximum number of CDNC pixels available within the $1^\circ \times 1^\circ$ domain for the selected overpass (shown with red dashed lines in the figures). The unfiltered data sets show almost full cloud coverage within the $1^\circ \times 1^\circ$ domains at both sites. Although this provides the largest spatial coverage, these pixels are not necessarily suitable for aerosol–cloud interaction analysis, because they may include retrievals affected by unsuitable viewing geometry, cloud heterogeneity, non-liquid cloud phase, or cloud structures that do not satisfy the assumptions required for CDNC retrieval. The general filters restrict the sample to more physically suitable liquid-cloud retrievals. When the general filters are used to select pixels that represent low-level water clouds and have high quality, the number of pixels drop to almost half in Hyytiälä and to one third in SGP. The local filter then removes pixels located near potentially problematic retrievals, particularly neighboring ice-cloud pixels or unsuccessful liquid-cloud retrievals. The addition of the local filter reduces the number of pixels by additional 2 % units in Hyytiälä and by 3 % units in SGP. Finally, following the adiabatic-consistency criterion described above, the sample was further restricted to



pixels satisfying $CER_{3.7} \geq CER_{2.1}$. The usage of the adiabatic filter reduces the number of pixels further by 12 % units in Hyytiälä and by 7 % units in SGP.

360 Together, these filters reduce the number of retained pixels but improve the physical consistency of the sampled cloud population. For the example overpasses shown in 5, approximately 41% of the initial pixels were retained over Hyytiälä and 22% over SGP after all filters were applied. These percentages are provided only to illustrate the effect of the filtering procedure for the selected cases and are not intended to represent retention fractions for the full dataset. Therefore, the final dataset should be interpreted as representing well-constrained low-level liquid-cloud conditions, improving the reliability of the CDNC
 365 estimates used in the aerosol–cloud susceptibility and radiative feedback analysis. All the following quantitative statistics, aerosol–cloud relationships, and radiative feedback estimates are derived from the complete set of overpasses satisfying these filtering criteria.

3.3 Aerosol-Cloud Susceptibility

Figure 6 shows the relationship between MODIS-derived $\ln(CDNC)$ and in-situ $\ln(N_{100})$ over the $1^\circ \times 1^\circ$ domains around
 370 Hyytiälä and SGP for non-adiabatic (panels a and c) and adiabatic (panels b and d) conditions. The slopes of these relationships represent the aerosol–cloud susceptibility, $\partial \ln(CDNC) / \partial \ln(N_{100})$. The figure compares three observational regression methods, OLS, YR and DR, together with OLS slopes based on cloud parcel model simulations. We used the YR and DR methods to account for uncertainty in both CDNC and N_{100} observations. For each dataset, the different fitting methods produce comparable slopes within their uncertainty ranges. This indicates that the inferred positive CDNC – N_{100} relationships are not
 375 strongly dependent on the choice of the regression method, and that measurement uncertainty does not fundamentally change the interpretation of the aerosol–cloud susceptibility. Furthermore, the slopes based on the parcel model results are also well in line with the observational estimates, especially for the adiabatic sampling conditions. This indicates that the observations are aligned with our theoretical understanding on the activation of aerosols in these regions.

For Hyytiälä, the aerosol–cloud relationship is clear both with and without the adiabatic constraint. Without the adiabatic
 380 criterion, the observational relationship is present (slopes of 0.29–0.32) but significantly smaller than what one would expect based on theory (slope of 0.52). After applying the adiabatic constraint, the relationship becomes stronger and more coherent, and the observational slopes increase to 0.42–0.46. The confidence intervals of these estimates do not overlap with the estimates without adiabatic criterion and they are mainly within the confidence interval of the model estimate. This indicates that the adiabatic liquid-cloud cases show a more robust CDNC response to the variability in N_{100} . This behavior suggests that part
 385 of the weaker relationship in the non-adiabatic sample may arise from non-ideal cloud scenes, entrainment effects, drizzle, or cloud heterogeneity. To assess whether the inferred aerosol–cloud susceptibility depends on the choice of spatial sampling domain, the same analysis was repeated for the 0.5° and 2° domains around Hyytiälä (see Supplementary Figure S4). The main behavior remains consistent across the domains. In both cases, the adiabatic samples give larger CDNC – N_{100} slopes than the corresponding non-adiabatic samples, despite the substantial reduction in the number of observations. The observed
 390 slopes in the non-adiabatic samples are generally smaller than the cloud parcel model slopes, whereas the adiabatic samples show closer agreement between observations and the model response. The observed adiabatic slopes remain similar across all

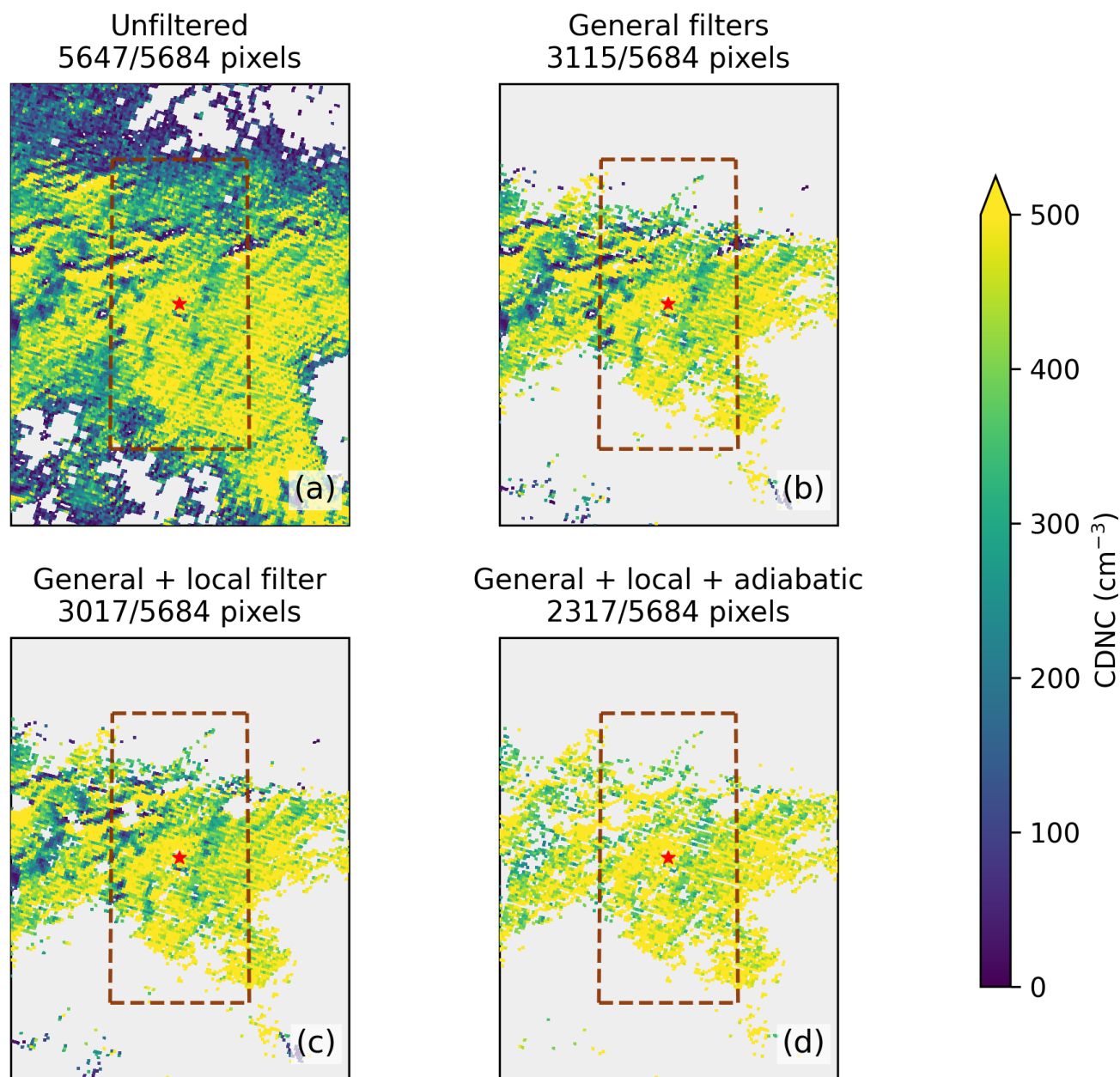


Figure 4. Terra MODIS Level-2 CDNC over Hyytiälä for the overpass on 23-08-2016, 09:45, showing the effect of successive sampling criteria within the $1^\circ \times 1^\circ$ domain. Panels show (a) unfiltered retrievals, (b) general cloud and viewing-geometry filtering, (c) local filtering, and (d) with adiabatic assumption. The red star indicates the site location, the dashed box shows the sampling domain, and the pixel counts indicate the retained in-domain pixels after each filtering step.

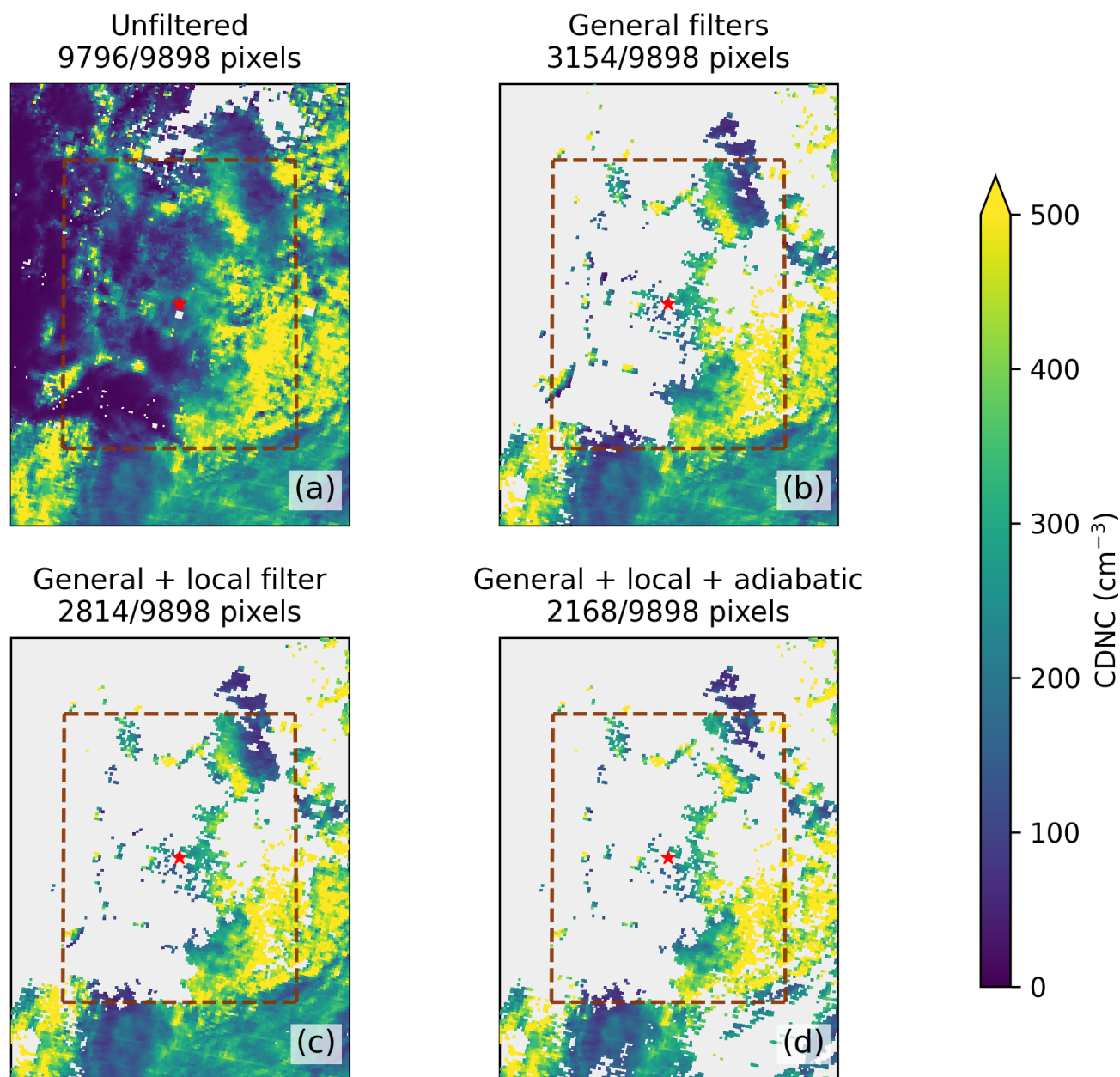


Figure 5. Terra MODIS Level-2 *CDNC* over SGP for the overpass on 01-06-2021, 17:20, showing the effect of successive sampling criteria within the $1^\circ \times 1^\circ$ domain. Panels show (a) unfiltered retrievals, (b) general cloud and viewing-geometry filtering, (c) local filtering, and (d) with adiabatic assumption. The red star indicates the site location, the dashed box shows the sampling domain, and the pixel counts indicate the retained in-domain pixels after each filtering step



domains, approximately 0.40–0.48, and the differences among regression methods are small. The cloud parcel model slopes also show only modest domain dependence, with a slightly lower value for the 0.5° domain than for the 1° and 2° domains. Overall, these supplementary analyses indicate that the Hyytiälä CDNC – N_{100} susceptibility is relatively robust to the choice of sampling domain, while the adiabatic cloud-selection criterion has a larger influence on the inferred slope.

For SGP, six observations were excluded after additional diagnostic screening. These cases showed large spatial variability in pixel-level CDNC within the MODIS sampling domain, with relative spatial standard deviations of approximately 25–58%, and in some cases the overpass-mean CDNC was strongly influenced by very large pixel-level CDNC values. They were therefore interpreted as poorly representative overpass-mean CDNC estimates and excluded from the analysis. Even with the screened data set, the CDNC – N_{100} relationship at SGP is not as clear as over Hyytiälä. This is likely due to the lower number of observations and the lack of N_{100} concentrations below 150 cm^{-3} ($\ln(N_{100}) < 5 \text{ cm}^{-3}$). The observational slopes for the dataset without adiabatic criteria (0.28–0.40) are in the same range as for Hyytiälä, but the confidence intervals are much larger. After applying the adiabatic constraint, the observed slopes increase to the range of 0.36–0.46 which is in the range of the confidence interval of the model based estimate.

The analysis for SGP was also performed for the 0.5° and 2° domains to assess the sensitivity of the CDNC – N_{100} relationship to spatial sampling (see Supplementary Figure S5). In contrast to Hyytiälä, the domain-size sensitivity at SGP does not show a consistent strengthening of the aerosol–cloud response under adiabatic sampling. For the 0.5° domain, the adiabatic slopes are lower than the corresponding non-adiabatic slopes, whereas for the 2° domain the adiabatic slopes are higher than the non-adiabatic slopes. The differences among OLS, YR, and DR slopes are generally small, suggesting that the broad interpretation is not strongly dependent on the regression method. However, the overall slopes remain lower than those found at Hyytiälä, and the agreement with the cloud parcel model is weaker, indicating that the aerosol–cloud susceptibility is less clearly constrained at SGP.

The cloud parcel model slopes are generally slightly steeper than the observational slopes, especially for Hyytiälä. This could indicate that the model predicts a stronger CDNC response to aerosol number than is observed in the satellite–in-situ collocations. The difference is expected because the model represents an idealized activation framework with prescribed aerosol modes and composition, whereas the observations include variability in updraft velocity, temperature, entrainment, cloud mixing state, aerosol composition, and retrieval uncertainty. However, the observational susceptibility slopes tend to fall within the uncertainty range of the cloud parcel model slope, indicating that the observed CDNC response to N_{100} is physically consistent with aerosol activation theory. The Figure 6 b) also shows a clear positive dependence of N_{100} on temperature over Hyytiälä (shown with the colors of the points). This supports the proposed boreal feedback pathway, consistent with enhanced biogenic emissions and secondary aerosol formation under warmer conditions. In contrast, no clear N_{100} –temperature dependence is observed at SGP. This suggests that N_{100} at SGP is not driven by temperature-dependent aerosol emissions. Instead, N_{100} is likely influenced by a broader combination of factors, including anthropogenic emissions, boundary-layer mixing, and variable synoptic conditions. As a result, the temperature-driven aerosol pathway is less clearly isolated at SGP than at Hyytiälä.

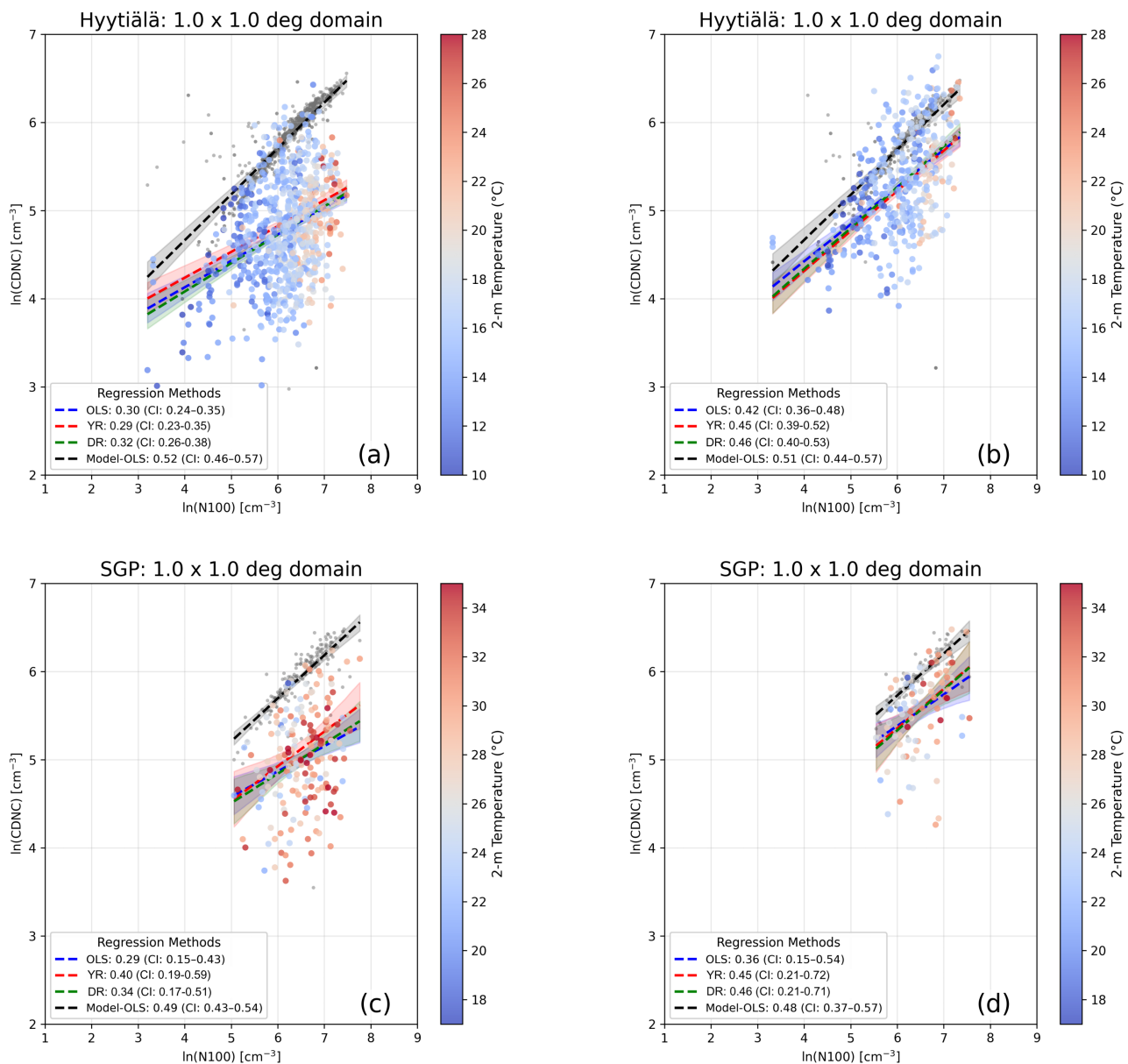


Figure 6. Aerosol–cloud susceptibility over the $1^\circ \times 1^\circ$ domains around Hyytiälä and SGP. Panels show collocated $\ln(\text{CDNC})$ from MODIS Level-2 retrievals versus in-situ $\ln(N_{100})$ for non-adiabatic (left-hand panels) and adiabatic sampling (right-hand panels) conditions: Hyytiälä (a, b) and SGP (c, d). Observational slopes are estimated using OLS, YR, and DR, with the cloud parcel model OLS slope overlaid for comparison. The slopes represent $\partial \ln(\text{CDNC}) / \partial \ln(N_{100})$, and shaded bands show 95% confidence intervals.



Table 1. Direct aerosol radiative feedback estimates for the $1^\circ \times 1^\circ$ domains around Hyytiälä and SGP under clear-sky and all-sky conditions. For SGP the estimate is based on the NO_2 -adjusted N_{100} -temperature coefficient from multivariate regression. Values are given with 95 % confidence intervals in parentheses. The clear-sky estimates represent the aerosol radiative response without cloud masking, whereas the all-sky estimates account for the reduction of the direct aerosol effect under cloudy conditions.

Direct Radiative Feedback Estimates ($\text{Wm}^{-2}\text{C}^{-1}$)		
Site	Clear-sky (95% CI)	All-sky (95% CI)
Hyytiälä	-0.56 (-0.72 to -0.39)	-0.27 (-0.35 to -0.19)
SGP (NO_2 -adjusted)	-0.28 (-0.32 to -0.23)	-0.14 (-0.16 to -0.12)

3.4 Direct and Indirect radiative feedback estimates

Table 1 summarizes the direct aerosol radiative feedback estimates for the $1^\circ \times 1^\circ$ domains at Hyytiälä and SGP. The feedback is negative at both sites under both clear-sky and all-sky conditions, indicating a cooling response associated with temperature-dependent changes in AOD. At Hyytiälä, the clear-sky feedback is -0.56 [CI: $-0.72, -0.39$] $\text{Wm}^{-2}\text{C}^{-1}$. The corresponding all-sky ($C_c=0.51$) value is smaller in magnitude, -0.27 $\text{Wm}^{-2}\text{C}^{-1}$, because cloud cover reduces the additional radiative effect of aerosol scattering. The direct feedback thus obtained for SGP is also negative but weaker than at Hyytiälä, with a clear-sky value of -0.28 [CI: $-0.32, -0.23$] $\text{Wm}^{-2}\text{C}^{-1}$ and an all-sky ($C_c=0.49$) value of -0.14 $\text{Wm}^{-2}\text{C}^{-1}$. For the $1^\circ \times 1^\circ$ domains, the clear-sky direct feedback is stronger at Hyytiälä than at SGP due to the longer path length of direct sunlight through the atmosphere (larger ϕ) and smaller surface reflectance at the boreal site.

Yli-Juuti et al. (2021) reported twice as high clear-sky direct radiative feedback for Hyytiälä (-1.15 $\text{Wm}^{-2}\text{C}^{-1}$ with 95 % credible interval from -2.49 to -0.47 $\text{Wm}^{-2}\text{C}^{-1}$). Their estimate is more negative due to a stronger AOD-temperature relationship (0.015 [CI: $0.006, 0.031$] $^\circ\text{C}^{-1}$) in their analysis but our estimate is within their uncertainty envelope. However, our confidence interval is narrower due to the longer time series, which implies that the direct effect in Hyytiälä is most likely around -0.5 $\text{Wm}^{-2}\text{C}^{-1}$. Moreover, our estimate for the direct aerosol radiative feedback at SGP is well in line with the estimate by Mielonen et al. (2018) (-0.33 ± 0.29 $\text{Wm}^{-2}\text{C}^{-1}$) for the southeastern USA. Consequently, the clear-sky direct radiative feedback caused by biogenic aerosols is most likely around -0.6 $\text{Wm}^{-2}\text{C}^{-1}$ in a clean environment and approximately half of that in a rural region.

Table 2 summarizes the indirect cloud-albedo radiative feedback estimates for the $1^\circ \times 1^\circ$ domains at Hyytiälä and SGP. The feedback is negative at both sites and under both sampling assumptions, indicating a cooling response associated with temperature-dependent aerosol changes and their effect on cloud droplet number concentration.

At Hyytiälä, the indirect feedback is -0.41 [CI: $-0.52, -0.31$] $\text{Wm}^{-2}\text{C}^{-1}$ without the adiabatic constraint and increases in magnitude to -0.58 [CI: $-0.71, -0.46$] $\text{Wm}^{-2}\text{C}^{-1}$ when the adiabatic criterion is applied. This increase is statistically significant and consistent with the stronger $\text{CDNC} - N_{100}$ susceptibility obtained for the adiabatic cloud case. This suggests that the aerosol influence on CDNC, and consequently on cloud albedo, is more clearly detectable when the analysis is restricted



Table 2. Indirect radiative feedback estimates for the $1^\circ \times 1^\circ$ domain around Hyytiälä and SGP. Values are reported with 95 % confidence intervals in parentheses. Estimates are shown with and without the adiabatic sampling criterion. For SGP, the estimate is based on the NO_2 -adjusted N_{100} -temperature coefficient from multivariate regression.

Indirect Radiative feedback ($\text{Wm}^{-2}\text{C}^{-1}$) - $1^\circ \times 1^\circ$ domain		
Site	With Adiabatic assumption	Without Adiabatic assumption
Hyytiälä	-0.58 (-0.71 to -0.46)	-0.41 (-0.52 to -0.31)
SGP (NO_2 -adjusted)	-0.81 (-1.25 to -0.31)	-0.63 (-0.97 to -0.30)

to liquid-cloud scenes that satisfy the retrieval-quality and adiabatic-consistency criteria. Yli-Juuti et al. (2021) estimated the indirect feedback in Hyytiälä to be -0.30 [CI: -0.58 , -0.04] $\text{Wm}^{-2}\text{C}^{-1}$ which is below our confidence interval when adiabatic criteria are used. If we do not use the adiabatic criteria, our estimate is closer to the one provided by them. They had used Level 3 MODIS cloud data, which does not consider the adiabaticity of the retrievals. Therefore, it seems that if adiabaticity is not considered, the satellite-based estimates of indirect cloud-albedo feedbacks are likely underestimated.

At SGP, the magnitude of the NO_2 -adjusted feedback is -0.63 [CI: -0.97 , -0.30] $\text{Wm}^{-2}\text{C}^{-1}$ without the adiabatic constraint, and -0.81 [CI: -1.25 , -0.31] $\text{Wm}^{-2}\text{C}^{-1}$ when the adiabatic criterion was applied. Unlike in Hyytiälä, the consideration of the adiabatic criteria does not introduce statistically significant differences between the estimates due to the wide confidence intervals. Furthermore, these values should be interpreted as rough estimates because the tropospheric NO_2 concentrations do not capture all anthropogenic, transport, boundary-layer, or removal processes influencing aerosol number. However, both estimates are within the model based approximation of effective radiative forcing for biogenic aerosols in southeastern US ($-1.0 \pm 0.5 \text{Wm}^{-2}\text{C}^{-1}$; Mielonen et al., 2018), which indicates that the indirect feedback in this region is likely in the vicinity of $-1.0 \text{Wm}^{-2}\text{C}^{-1}$.

The increase in feedback magnitude under the adiabatic assumption at both sites highlights the sensitivity of the indirect feedback estimate to cloud sampling strategy. The adiabatic criterion selects clouds with a more physically plausible vertical droplet-size structure, for which satellite CDNC retrievals are expected to be more reliable. Non-adiabatic or heterogeneous cloud scenes may dilute the aerosol signal through entrainment, drizzle, cloud-edge effects, or retrieval uncertainty. Therefore, the stronger adiabatic feedbacks suggest that strict cloud sampling reveals a more coherent aerosol–cloud response.

Overall, the magnitude of radiative feedback is sensitive to cloud sampling and to the definition of the aerosol temperature response. At Hyytiälä, the indirect feedback is comparable in magnitude to the direct clear-sky aerosol feedback, indicating that indirect cloud-albedo effects are an important component of the boreal aerosol feedback. At SGP, the NO_2 -adjusted multivariate estimate is approximately three-times larger than the direct clear-sky aerosol feedback, indicating a much stronger indirect effect and that the direct and indirect effects have different relationships in different environments.



4 Conclusions

We have quantified temperature-dependent aerosol radiative feedbacks over two distinct land environments, the boreal forest site Hyytiälä and the continental SGP site, by combining MODIS Level-2 cloud retrievals, in-situ aerosol number concentrations, meteorological observations, and cloud parcel model simulations. The analysis was designed to connect aerosol changes with cloud droplet number concentration, cloud albedo, and radiative feedback, while explicitly testing the sensitivity of the results to MODIS cloud sampling assumptions and fitting methods.

The MODIS sampling analysis shows that successive filtering substantially reduces the number of available cloud pixels, but provides a more physically consistent subset of low-level liquid-cloud retrievals. The final filtered samples exclude pixels affected by unsuitable viewing geometry, non-liquid or multilayer cloud conditions, local retrieval inconsistencies, and non-adiabatic cloud structures. Therefore, stricter sampling reduces data availability but improves the physical interpretability of the COT and CER retrievals used in the calculation of CDNCs and the following aerosol–cloud interaction analysis.

Positive CDNC – N_{100} relationships were found at both sites with and without the adiabatic constraint, indicating that aerosol variability has an observable contribution to cloud droplet number variability under the selected liquid-cloud conditions. At both sites, the aerosol–cloud relationship becomes stronger after applying the adiabatic criterion. This suggests that the aerosol influence on CDNC is more clearly detectable when the analysis is restricted to physically consistent liquid-cloud scenes.

The comparison of OLS, YR, and DR slopes showed that accounting for uncertainty in both CDNC and N_{100} , including satellite retrieval uncertainty and spatial variability in CDNC, does not substantially change the inferred aerosol–cloud susceptibility. This indicates that the positive CDNC – N_{100} response is robust and not dependent on the regression method.

The clear-sky direct radiative feedback caused by biogenic aerosols is most likely around $-0.6 \text{ W m}^{-2} \text{ °C}^{-1}$ in a clean environment and approximately half of that in a rural region. The all-sky radiative feedbacks are half of the clear-sky estimates at both locations. The indirect radiative feedbacks of biogenic aerosols are in the same range or up to three times larger than their direct radiative feedbacks depending on the region. This makes the quantification of the global climate effects of biogenic aerosols challenging.

Overall, our analysis shows that biogenic aerosols introduce significant climate feedbacks. However, they are likely underestimated if the satellite-based cloud retrievals are not properly sampled. Based on our results, the analysis should be restricted to well-resolved single-layer liquid clouds, exclude cloud-edge pixels and retrievals adjacent to failed or ice-contaminated pixels, and apply an adiabatic-consistency criterion to retain cloud scenes with vertical variations in droplet size. These sampling steps reduce the number of available observations but strengthen the retrieved aerosol–cloud relationship and improve the robustness of the resulting radiative feedback estimates. On the other hand, the estimation of cloud susceptibility is not sensitive to the selection of the fitting method. Furthermore, we show that state-of-the-art droplet formation parameterizations are able to reproduce observations, thus cloud parcel modeling can be used to evaluate the robustness of observational results. These findings demonstrate the potential of combining carefully sampled satellite retrievals, in-situ aerosol observations, and process modelling to improve the estimates of aerosol-cloud radiative feedbacks over land.



Code availability. The Python codes used in the analysis and production of the figures are available at <https://doi.org/10.57707/fmi-b2share.hdb6g-g7n68>.

Data availability. Field data on particle size distributions from Hyytiää are available at <https://smear.avaa.csc.fi/download>. ECMWF-ERA5 data are available at the Copernicus Climate Change Service (C3S) Climate Data Store <https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5> (Hersbach et al., 2023). The Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) provided the CLU (2023) dataset in this study (CLU, 2023). CLU data was produced by the Finnish Meteorological Institute (FMI) and is available for download from <https://cloudnet.fmi.fi/>. The aerosol data for Southern Great Plains were obtained from the Atmospheric Radiation Measurement (ARM) User Facility, a U.S. Department of Energy (DOE) Office of Science user facility managed by the Biological and Environmental Research Program. AERONET data are available at <https://aeronet.gsfc.nasa.gov/>. MODIS Level-2 cloud products were obtained from the NASA Level-1 and Atmosphere Archive and Distribution System Distributed Active Archive Center (LAADS DAAC), and OMI tropospheric (NO₂) data were obtained from the NASA Goddard Earth Sciences Data and Information Services Center (GES DISC).

Author contributions. T. M., K. T., S. R., H. K., and A. A. designed the study. T. P., M. K., S. K. provided the field measurements. K. T., T. M., S. C., S. R., H. K., A. A., S. C., and A. L. processed and analyzed the data. K. T. and T. M. wrote the manuscript with input from all the authors.

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