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Suzanne Crumeyrolle, Andrey Khlystov, Harry ten Brink. On the Trend in Below-Cloud Solar Irradiance in The Netherlands versus That in Aerosol Sulfate Concentration. *Atmosphere*, 2023, *Atmosphere*, 13, 10.3390/atmos13122037 . hal-04459236

HAL Id: hal-04459236

<https://hal.univ-lille.fr/hal-04459236>

Submitted on 15 Feb 2024

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Article

On the Trend in Below-Cloud Solar Irradiance in The Netherlands versus That in Aerosol Sulfate Concentration

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Abstract: The below-cloud irradiance in The Netherlands increased by over 10 W m^{-2} in the past half century. It was hypothesized that this could be due to a decrease in the aerosol serving as cloud condensation nuclei, on which the cloud droplets form, in the following way. With unchanged macrophysics, clouds with a lower number of droplets are less reflective, in other words, they transmit more solar radiation. This hypothesis cannot be substantiated with data because of a generic absence of long-term information on cloud droplet number concentrations (CDNCs) worldwide. To assess the historic trend in CDNC, we used the Boucher and Lohmann (B&L) empirical relationship between CDNC and the mass concentration of the water-attracting hygroscopic aerosol components. The B&L parameterization was tested and validated with observations from the CHIEF cloud chamber, in which the formation of marine stratocumulus, the most frequent cloud type in The Netherlands, is simulated. This study will focus on summer periods because the irradiance governs the yearly average at this latitude. The summer trend of sulfate, the most dominant hygroscopic component of observed aerosol mass concentration, was analyzed with EBAS data from 1972 onwards. The average summer CDNCs were then calculated via the B&L parameterization, showing an upper limit of 380 cm^{-3} in the 1970s and decreasing to around 200 cm^{-3} in the 2010s. The associated increase in transmission for thin marine stratocumulus without overlying clouds would be, at most, 3.5 W m^{-2} . Unobstructed stratocumuli occur only part of the time, and the change in irradiance based on the reduction in cloud droplet number is certainly small in comparison to the empirically derived trend of 10 W m^{-2} .

Keywords: aerosol–cloud interaction; cloud chamber; stratocumulus

Citation: Crumeyrolle, S.; Khlystov, A.; Ten Brink, H. On the Trend in Below-Cloud Solar Irradiance in The Netherlands versus That in Aerosol Sulfate Concentration. *Atmosphere* **2022**, *13*, 2037. <https://doi.org/10.3390/atmos13122037>

Academic Editors: Wenhao Xue, Jing Zhang, Jing Wei and Yulu Tian

Received: 28 September 2022

Accepted: 28 November 2022

Published: 4 December 2022

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1. Introduction

Aerosols have a significant impact on the Earth's radiation budget by scattering and absorbing solar radiation (direct effect), leading to negative (cooling) and positive (warming) radiative forcing. Moreover, interactions between aerosols and clouds (indirect effect) have a large influence on the role of aerosols on climate change. The atmospheric aerosols affect cloud formation through their role as cloud condensation nuclei (CCN). Sulfate aerosols are nowadays considered as the most dominant hygroscopic component of aerosols [1,2]. Sulfate is an important secondary aerosol formed from photochemical reactions of sulfur dioxide (SO_2) emissions in the atmosphere. Summertime peaks of sulfate concentrations are common for most areas, including The Netherlands [3], due to available solar insolation, chemical reactions facilitated in high relative humidity environments, and stagnation events e.g., [4,5]. To reduce SO_2 emissions over the world, regulatory and legislative mandates were established and total annual SO_2 emissions over Europe have decreased by 72% from 1980 to 2004 [6,7], and by 60% from 1990 to 2010 over the United States [8].

Over the past 50 years, the solar irradiance over The Netherlands significantly increased, both during clear-sky and cloudy situations [9]. The solar irradiance increase

during clear sky conditions could be attributed to a decrease in reflective aerosols within the atmosphere, such as sulfate [10,11]. Whereas, the solar irradiance decrease under cloudy conditions may have been caused by a decrease in CDNC, leading to a reduction in cloud reflectivity [12–15]. Assuming a constant liquid water path (LWP), a decrease in CDNC would be due to a lower CCN concentration. In this case, CDNC and below-cloud irradiance trends should be similar.

Over the North Sea region, stratocumulus (SC) is the most common [16] and the most frequent cloud type. Together with stratus, it covers the North Sea nearly 50% of the time [16]. SC generally forms at the top of the planetary boundary layer, and is therefore connected to aerosol observed at the surface. Moreover, previous studies, based on remote sensing, parcel modeling, and large eddy simulation (LES), have highlighted that SC clouds exhibit substantial albedo susceptibility to variations in CDNC [17,18].

Trend analyses are needed to track the evolution of solar radiation and CDNC. In a trend analysis, the annual average of the solar irradiance measured values cannot be used because of its variation, which depends on the solar angle and cloud coverage. For these reasons, cloud cover values were scaled to the yearly average of cloud cover and occurrence of clouds per solar angle [9]. The parameter obtained is defined as a “proxy” for below-cloud irradiance per year. At The Netherlands latitudes, the annual solar irradiance is determined by that in summer because of the long sunshine hours and high irradiance. This leads to a difference of a factor of ten in irradiance in summer and winter [19].

Contrary to solar irradiation, the CDNC trend cannot be evaluated from measurements because of an absence of monitoring data. This is not a local issue but a worldwide phenomenon. To assess the historic CDNC trend, de Martino et al. [20] detailed several approaches, including using the mass concentration of the hygroscopic aerosol components as a proxy. In a recent evaluation of the largest dataset in marine clouds in South California [21], the parameterization of Boucher and Lohmann (B&L) [22], based on the mass concentration of sulfate to retrieve CDNC, was found to provide the best results in comparison to the data. This was also the case in a shorter study off the Canadian coast of the North Atlantic [23].

There is very limited information on the CDNC over the North Sea region, let alone in combination with sulfate concentrations. Only some data exist on the CDNC in the 1980s [24], mostly collected in continental air masses non-representative of the background concentrations. In view of this, we derived an evaluation of the relationship between CDNC and sulfate concentration in a large cloud chamber (CHIEF) at the northwestern seacoast of The Netherlands, in which the conditions of marine stratocumulus were simulated [25].

In the present investigation, we first tested whether the B&L parameterization was appropriate for the relevant marine clouds over The Netherlands via an investigation in a large cloud chamber (CHIEF). Then, the summer trend of the sulfate concentration was reconstructed using EBAS data to infer the CDNC trend using the B&L parameterization. Finally, the CDNC trend was used to estimate the modification in solar irradiance only due to indirect effect, and will be compared to below-cloud irradiance trend. This will provide information on the magnitude of the aerosol–cloud interaction (ACI) effect on climate [3]. Before we proceed, it is important to mention that in addition to sulfate, other chemical compounds in aerosols, such as nitrate [20], might be of importance as components of the cloud nuclei. However, we showed in a recent comprehensive study [25] that, in The Netherlands, ammonium nitrate is not relevant during the summertime, and neither are organic compounds or sea salt; even though they significantly contribute to the total aerosol mass, they are not present in the size range of particles that serve as cloud nuclei.

2. Materials and Methods

2.1. Cloud Chamber

The large cloud chamber (CHIEF) used in the tests is situated at the northwestern seacoast of The Netherlands. A full description of the set-up is provided in an earlier publication [25]. Here we provide a summary of the relevant aspects. It is a 21 m³ flow-

through chamber in which the low supersaturation conditions of marine stratocumulus are simulated [25]. The proper setting of the chamber was derived from simultaneous CDNC measurements within the chamber and aboard a small aircraft flying in stratocumulus over the site. These results are detailed for the first time in Appendix A. The CDNC was measured using a forward scattering spectrometer probe (FSSP-100 from DMT) for droplets within the diameter range of 3 to 47 μm . Size and number concentration were calibrated according to the guidelines in the manual. The sulfate concentration was observed with a MARGA-sizer that works as follows: particles are grown to large droplets by steam condensation, and these are collected in a cyclone [26,27]. In the drain water, the hygroscopic species dissolve and are then measured online as ions. The accuracy of the MARGA system has been checked against 24-h filter measurements, e.g., [28,29], with an average deviation of less than 3%.

2.2. Historic Sulfate Measurements

The sulfate mass concentration has been measured since July 1972 in The Netherlands, with many of the data included in the EMEP/EBAS database [30]. A previous study [31] already conducted an analysis comparing the trend of observed sulfate mass concentration to the reflection of radiation during summer over the years 1978–1994. Since the publication of this study, the database has evolved to homogenize the mass concentration values as a function of the method used to measure it. In the early years, sulfate concentrations were measured through X-ray fluorescence (XRF), detecting sulfur, while in the period 1984–1987, a colorization reaction of the leached filter was applied [31]. Thereafter, ion chromatography was used, though data for the first 4 years of application of the latter method are not in the EBAS database. In 2009, PM_{10} samplers were introduced within the national network. An issue with this is that these collect a larger fraction of sea salt than the original samplers. To correct this bias, the PM_{10} samples were also analyzed for sodium, which allows the estimation of the sea salt sulfate fraction.

3. Results and Discussion

3.1. Cloud Chamber Study

Figure 1 shows the total sulfate concentrations and CDNC formed within the cloud chamber CHIEF during an intensive observation period in summer 2007. One can see that sulfate mass concentration ranges from 0.5 to 6 $\mu\text{g m}^{-3}$, while the corresponding CDNC (only) spans from 180 to 400 cm^{-3} . The large variability of the sulfate mass concentration is clearly not observed within the CDNC time series. Indeed, the CDNC did not significantly increase for sulfate concentrations exceeding 3 $\mu\text{g m}^{-3}$. However, one can see that the tendency is similar in both figures, with two large peaks observed on the 1st and on 12 August 2007 for both parameters. On the 5 August 2007, the CDNC peak reached up to 366 cm^{-3} , while the sulfate was unfortunately not measured due to technical issues. However, the sulfate concentration before and after this date is rather large ($>3 \mu\text{g cm}^{-3}$), probably suggesting a peak on the 5 August.

As for the functional relationship of the CDNC and the mass concentration, we refer to Figure 2 (blue cross), in which the relationship of Leitch et al. [23] is added. These data derive from the concentration of sulfate in the cloud water translated to the corresponding air concentration. For this translation, the liquid water content (LWC) is used, for which the average uncertainty is 20%, as we deduce from the information given in the publication. This constitutes by far the largest measurement uncertainty. The Boucher and Lohmann (B&L) parameterization (dashed lines) for this relationship is provided, which thus holds for polluted coastal aerosol at the northwestern North Atlantic coast. It should be mentioned that the asymptotic value of the CDNC at the highest concentration agrees with that measured by Martin and Johnson [24] in the North Sea area (CDNC $\sim 350\text{--}400 \text{ cm}^{-3}$), substantiating the choice for the chosen parameterization. It seems that the observed CDNC in the cloud chamber is larger than the B&L parameterization. This can be explained by the fact that CHIEF, with its residence time of 2 min, simulates the situation at the cloud

base, where the CDNC is higher up in the cloud, as found in our cloud flight presented in Appendix A and by Leitch et al. [23]. Hence, CDNCs measured within the chamber are indeed expected to be higher than B&L retrievals. Incidentally, the presence of a higher concentration at the cloud base is of little relevance for the reflectivity, because it concerns very small droplets that evaporate upon further upward transport.

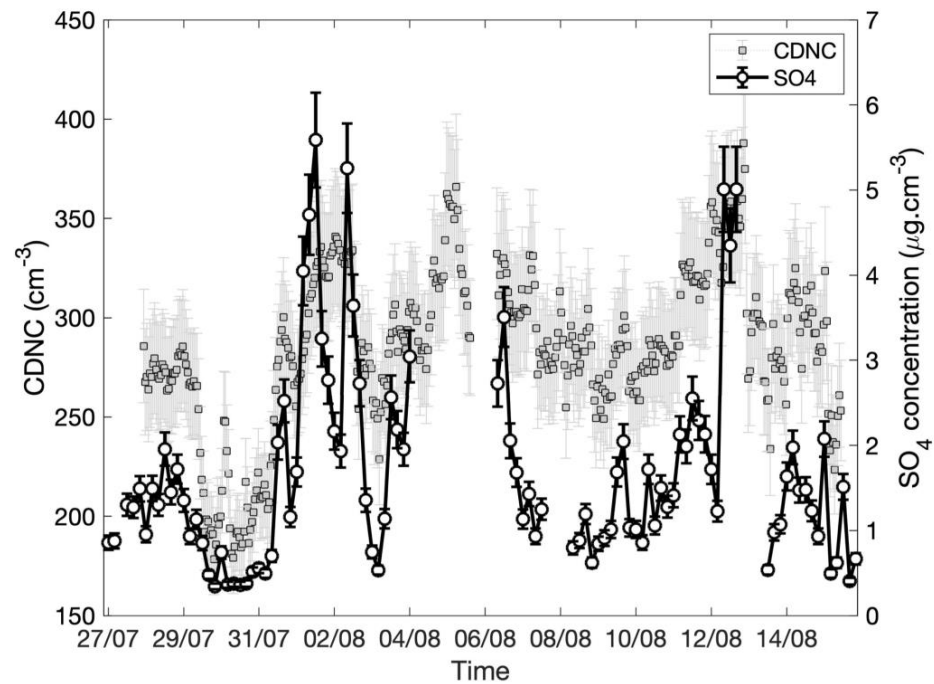


Figure 1. Sulfate mass concentration and cloud droplet number concentration (CDNC) measured within the cloud chamber (CHIEF) during the indicated intensive operational period in summer 2007.

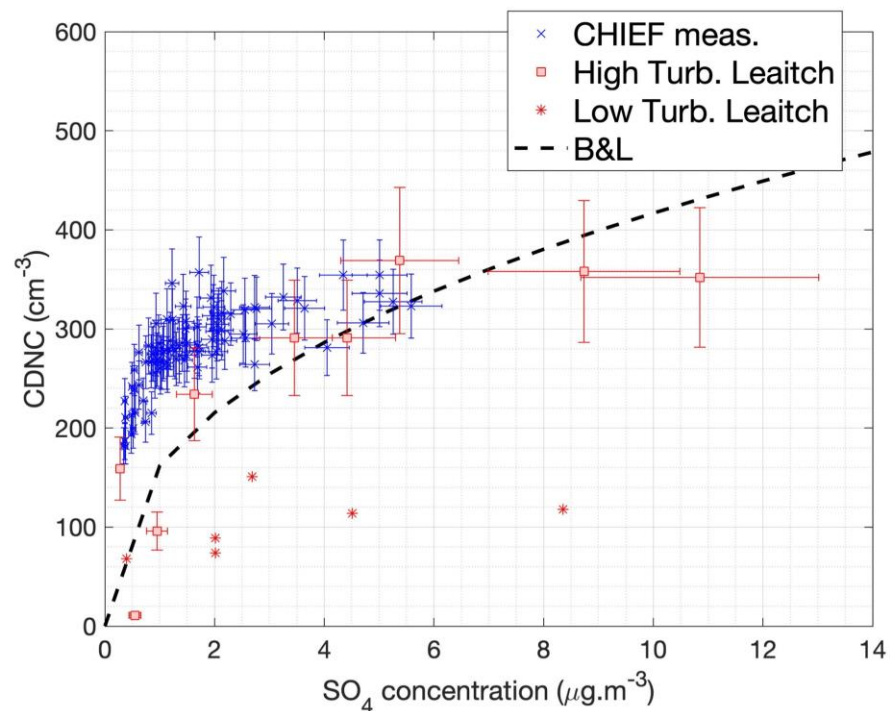


Figure 2. CDNC as a function of sulfate concentration measured in the CHIEF cloud chamber (blue cross) and observed during the stratocumulus flights of Leitch et al. [23] (red squares and stars). The dashed line represents the Boucher and Lohmann [22] parameterization.

As the CDNC follows the B&L parameterization rather well, the B&L parameterization could be used for the CDNC trend over The Netherlands. Therefore, in the next sections, the B&L parameterization will be used to estimate the trend in CDNC based on measured sulfate mass concentrations over the past ~50 years. It is also interesting to note, in Figure 2, that the maximum CDNC values observed within the CHIEF chamber are close to 350 cm^{-3} for sulfate concentrations larger than $1 \mu\text{g m}^{-3}$. This upper limit can also be seen in Leitch data, with a maximum observed at 380 cm^{-3} .

3.2. Sulfate Mass Concentration and CDNC Trends

All in all, non-sea-salt (nss) sulfate mass concentrations are available for eleven sites operational at different periods, with relatively often only one location active (see supplementary material for more information). There is a good correlation of data simultaneously collected at the different locations (Figure S2), which is consistent with the previous analysis [31] for The Netherlands in the 1980s and for stations, as far apart as 200 km, in western Europe in general [32]. Median nss-sulfate mass concentration per summer from only three sites (Witteveen, Appelscha, Vredepeel) were used to reconstruct the sulfate trend presented in Figure 3a. One can clearly see that the nss-sulfate trend is clearly decreasing with time. The median summer concentration in 1972 was around $10.2 \mu\text{g m}^{-3}$, and decreased down to $1.9 \mu\text{g m}^{-3}$ in 2019. This decrease is essentially due to stakeholder decisions at the European scale, especially the gasoline restrictions [10]. Moreover, one can clearly see that the daily average variability (grey dots) is also decreasing with time due to the decrease in daily PM exceedances also due to stakeholder decisions. A similar trend is observed over the southern part of the UK. For the years 1966 to 1971, the average summer concentration of nss-sulfate in the southern UK based on data at Chilton is close to $7 \mu\text{g m}^{-3}$ [33]. Currently (2016–2021), the concentration of nss-sulfate, observed in nearby Chilbolton, is just over $2 \mu\text{g m}^{-3}$.

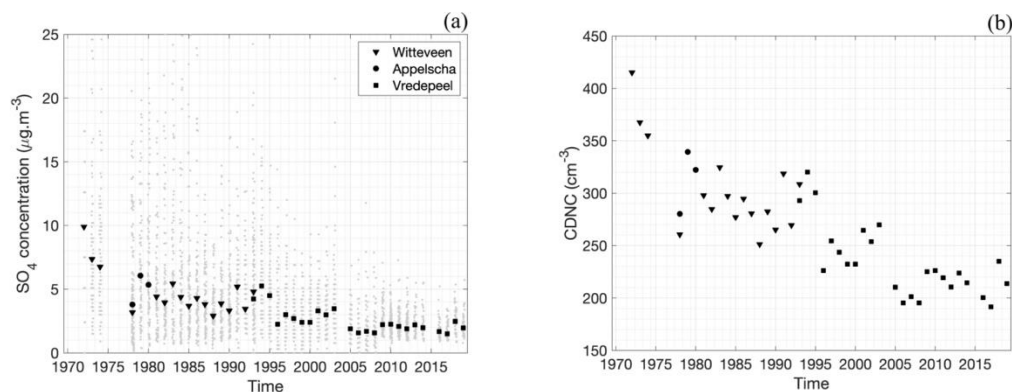


Figure 3. (a) Time series of non-sea-salt (nss) sulfate mass concentration observed over The Netherlands during the summer season (June–August) from three sites (Witteveen, triangles; Appelscha, dots; Vredepeel, squares; see supplementary material for more details). The black signs correspond to the median over the summer season and the grey signs to the daily averages. (b) Time series of CDNC derived from the Boucher and Lohmann relationship [22] applied to the nss-sulfate concentration shown in (a).

To estimate the summer CDNC trend, the relationship (B&L (A) and (D), shown in Figure 2) between CDNC and nss-sulfate mass concentration is used (Figure 3b). With a current-day nss-sulfate concentration of around $1.9 \mu\text{g m}^{-3}$ over The Netherlands, the CDNC should be in the order of 200 cm^{-3} . Meanwhile, in the 1970s, the nss-sulfate concentration was over $10 \mu\text{g m}^{-3}$ and the CDNC was retrieved at or even over the asymptotic maximum value of 380 cm^{-3} . A clear CDNC decrease, by a ratio up to 2, is observed during the last 40 years.

The trend in the measured average annual irradiance [9] is reproduced in Figure 4a. From a plateau at $76 \pm 3 \text{ W m}^{-2}$ during the period 1966–1996, it increased to a level of

approximately $87 \pm 2.5 \text{ W m}^{-2}$ during the period 2000–2015. This difference (increase by 16%) is significant and far above the uncertainties. Therefore, the below-cloud solar radiation is clearly increasing over The Netherlands. As the CDNC is decreasing during the same period, the hypothesis of lower reflectivity clouds due to lower CDNC could explain this tendency. The cloud transmission would be larger, and then the total radiation below the cloud would increase as well.

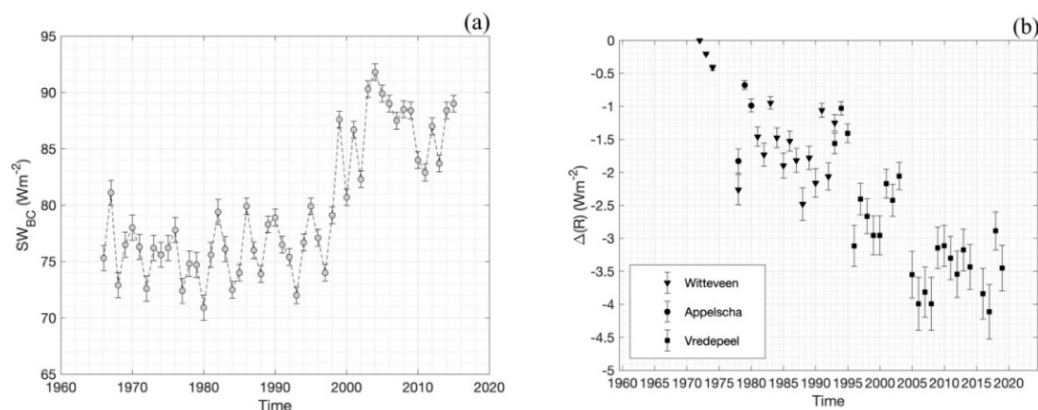


Figure 4. (a) Absolute cloud base proxy radiation as described by Boers et al. [9] over the indicated years averaged for five measuring sites. (b) Change in solar irradiance as deduced from the decrease in CDNC (Figure 3b).

In order to estimate this effect, we used the relationship provided by Schwartz and Slingo [34]. They established (as demonstrated in Equation (1)), based on older studies [31,35], that the decrease in reflectivity (ΔR) is associated with CDNC decrease considering a constant liquid water path (LWP) and cloud thickness (Z_c). This equation is accurate within 10% for the range of cloud-top albedo, $0.28 < R < 0.72$, characteristic of the prevalent and climatically important marine stratus clouds. It should also be noted that this relationship applies in the absence of overlying higher clouds. As the cloud profiles are not observed, we assume that there are only unobstructed stratocumulus clouds over The Netherlands, which means that the calculated reflectivity evolution is an upper limit.

$$(\Delta R)_{LWP, Z_c} = 0.075 * \Delta \ln(\text{CDNC}) \quad (1)$$

Assuming a constant LWP and cloud thickness throughout the last 50 summers over The Netherlands, the estimated SC reflectivity decreases by 3.5 Wm^{-2} in 2019 due to the decrease in CDNC. To add some perspective regarding the indirect forcing retrieved here, the total greenhouse gas forcing over the industrial period was about 2.5 W m^{-2} , with CO_2 contributing 1.5 W m^{-2} [36]. The indirect forcing on cloud reflectivity is therefore not negligible. As previously stated, this value is an upper estimate, given the fact that SC clouds observed over The Netherlands are not permanently unobstructed. Moreover, this estimated value is far below the observed increase in the below-cloud irradiance (around 10 Wm^{-2}). Therefore, the increase in the below-cloud irradiance can only be partly explained by the aerosol and cloud interaction in the lower layers. The rest of the increase could be due to a decrease in LWP and/or a decrease of cloud depth. Another possibility might be a systematic change in cloudiness values as a result of switching from a visual appraisal to objective probing by ceilometers [9]. This latter hypothesis could be checked via data for neighboring countries in which a similar trend in the mass concentration of sulfate occurred, but where cloudiness was very likely appreciated in a different manner over the years as compared to in The Netherlands.

4. Conclusions

In this study, Boucher and Lohmann parameterizations [22] were tested using cloud chamber (CHIEF) measurements of non-sea-salt (nss) sulfate mass concentration and cloud

droplet number concentration (CDNC). As expected, the CDNC measured in CHIEF, mimicking the cloud formation at its base, is overestimated in comparison to the B&L retrievals. Despite this overestimation, the results highlight that the B&L retrievals are reasonably comparable to our observations. The CDNC trend is then estimated from the B&L parameterizations, and the nss-sulfate mass concentration observed over The Netherlands for the past 50 years. Both the nss-sulfate and the CDNC trends are decreasing over the years. The CDNC was around 380 cm^{-3} in the 1970s, and decreased down to 200 cm^{-3} , while the nss-sulfate mass concentration has decreased by a factor of five over the last 50 years (from 10 to $2 \mu\text{g m}^{-3}$).

The CDNC evolution was then used to estimate the trend in stratocumulus cloud transmissivity. The below-cloud irradiance estimation increases by 3.5 W m^{-2} , at most, due to the presence of less CDNC. This increase can only explain part (35%) of the observed increase in below-cloud solar irradiance of 10 W m^{-2} . Therefore, there must be other parameters, such as cloud density or the appreciation of cloudiness over the years, that would explain this large modification of the solar irradiance received at the surface during cloudy days.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/atmos13122037/s1>, Figure S1: Sulfate monitoring sites listed in EBAS for The Netherlands (Table S1). The grey dots correspond to the sites used to build the sulfate long term trend. The diamond corresponds to Cabauw., Table S1: Netherland sites listed in EBAS data platform and the associated instrumentation used to measure sulfate. Table S2: Sulfate concentration ($\mu\text{g m}^{-3}$) measured at EBAS-NL sites (Table S1) since 1972. Figure S2: Comparison of sulfate concentration ($\mu\text{g m}^{-3}$) measured at different EBAS-NL sites (Table S1). The dashed line corresponds to the 1:1 line. Figure S3: Vertical profiles of LWC in the vicinity of Petten. Line indicates the ideal adiabatic relation between LWC and altitude.

Author Contributions: Conceptualization, H.T.B., A.K. and S.C.; methodology, H.T.B. and A.K.; formal analysis, H.T.B. and A.K.; investigation, H.T.B. and S.C.; writing—original draft preparation, H.T.B. and S.C.; writing—review and editing, H.T.B. and S.C. All authors have read and agreed to the published version of the manuscript.

Funding: The first author has been financially supported by the CaPPA project (Chemical and Physical Properties of the Atmosphere), which is funded by the French National Research Agency (ANR) through the PIA (Programme d'Investissement d'Avenir) under contract "ANR-11-LABX-0005-01".

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Sulfate measurements over The Netherlands are available through the EBAS database (<https://ebas.nilu.no>, accessed on 9 November 2021).

Acknowledgments: This work is a contribution to the CaPPA project (Chemical and Physical Properties of the Atmosphere), funded by the ANR through the PIA (Programme d'Investissement d'Avenir) under contract ANR-11-LABX-0005-01.

Conflicts of Interest: The authors declare no conflict of interest.

Appendix A. Cloud Base and Cloud Chamber CDNC Measurements

We present here the set-up and results of an aircraft study in low clouds over the site of Petten, where the CHIEF cloud chamber is located. The aim was to assess at which setting of the chamber a similar CDNC is observed as in the cloud layer above. The campaign took place in September 1996 with specific evaluation of the flight on September 4th with a closed cloud deck. The research aircraft was a PA 31 Navajo Chieftain two engine turbo-prop. Several horizontal transects were constructed in which the altitude was kept constant. The droplet number concentration was measured with a FSSP-100 (PMS Inc., Las Cruces, NM, USA), working at a frequency of 2 Hz, similar to that installed in the cloud chamber.

The aircraft encountered a closed cloud deck that had entered The Netherlands from the north. Surface winds were from the NNE for the entire period of the measurements (6:00 to 9:00 UTC). The back trajectory indicated that the air masses were mostly coming from Arctic marine regions. The cloud layer was capped by an inversion at about 1100 m according to sounding at a nearby airport. There was no observable precipitation.

Several horizontal tracks were constructed within the cloud in which the droplet number and size distribution were determined. From these measurements, the LWC and effective diameter (D_{eff}) were derived. The standard deviation of both parameters within one track was rather small (see Table A1). The droplet spectrum shifts towards larger sizes at higher altitudes due to the continuing growth of the cloud droplets. The vertical profile of the LWC based on the droplet spectrum measured by the FSSP in the various horizontal tracks in the layer is shown in Figure S3. The experimental vertical profile of the LWC agrees reasonably well with the adiabatic profile of an almost ideal cloud layer.

One can see that the droplet number concentration (N) is relatively constant over the cloud height, apart from that measured at cloud base. Indeed, the CDNC value at cloud base (210 cm^{-3}) is higher than that higher up in the cloud layer. This observation is similar to that of Martin and Johnson [24] within SC over the same region. The cloud base measurements (862 m) are then comparable to the onset of the cloud formation as simulated in the CHIEF chamber.

Operating the CHIEF chamber in scanning mode [37], the supersaturation is ramped up and down in a cycle of 40 min. The aerosol spectrum was monitored with an SMPS, while the droplet spectrum was assessed with the same type of FSSP as on board the measuring aircraft flying above.

At the time of the overhead flight, the aerosol number concentration was 2800 cm^{-3} . The aerosol had a bimodal size distribution with an Aitken mode ($D_p = 50 \text{ nm}$) and an accumulation mode ($D_p = 150 \text{ nm}$). The best agreement of CDNC measured at cloud base (210 cm^{-3}) and within the cloud chamber was around a supersaturation of 0.1% (Figure A1). For further details, we refer to the detailed report by Khlystov et al. [38].

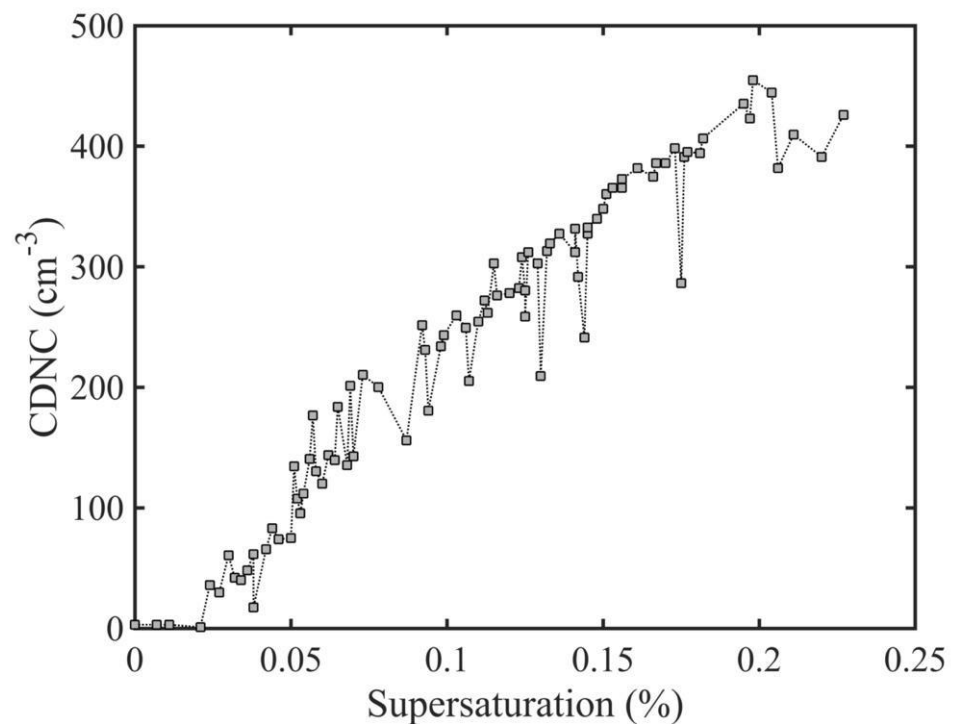


Figure A1. CDNC measured within the CHIEF cloud chamber as a function of supersaturation.

Table A1. Average and standard deviation (STD) of droplet number concentration (N), with LWC and effective diameter (D_{eff}) derived from the indicated horizontal tracks through a SC field in the vicinity of the CHIEF cloud chamber. Grey background is used for data measured at cloud base with higher variability in D_{eff} .

Altitude		N (# cm ⁻³)		LWC (mg m ⁻³)		D_{eff} (μm)	
Average	STD	Average	STD	Average	STD	Average	STD
862	11	210	60	170	60	6.6	1.5
895	3	110	35	250	50	18.8	1.3
895	3	90	20	250	50	19.4	1.0
942	26	145	70	420	165	19.9	0.6
970	5	120	30	530	60	22.1	0.9
1020	20	115	30	730	110	24.4	0.7
1025	5	120	40	650	145	23.5	0.9

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