



Spaceborne Observations of Lightning NO₂ in the Arctic

Environmental Science and Technology

Zhang, Xin; van der A, Ronald; Ding, Jieying; Eskes, Henk; van Geffen, Jos et al

<https://doi.org/10.1021/acs.est.2c07988>

This publication is made publicly available in the institutional repository of Wageningen University and Research, under the terms of article 25fa of the Dutch Copyright Act, also known as the Amendment Taverne.

Article 25fa states that the author of a short scientific work funded either wholly or partially by Dutch public funds is entitled to make that work publicly available for no consideration following a reasonable period of time after the work was first published, provided that clear reference is made to the source of the first publication of the work.

This publication is distributed using the principles as determined in the Association of Universities in the Netherlands (VSNU) 'Article 25fa implementation' project. According to these principles research outputs of researchers employed by Dutch Universities that comply with the legal requirements of Article 25fa of the Dutch Copyright Act are distributed online and free of cost or other barriers in institutional repositories. Research outputs are distributed six months after their first online publication in the original published version and with proper attribution to the source of the original publication.

You are permitted to download and use the publication for personal purposes. All rights remain with the author(s) and / or copyright owner(s) of this work. Any use of the publication or parts of it other than authorised under article 25fa of the Dutch Copyright act is prohibited. Wageningen University & Research and the author(s) of this publication shall not be held responsible or liable for any damages resulting from your (re)use of this publication.

For questions regarding the public availability of this publication please contact
openaccess.library@wur.nl

Spaceborne Observations of Lightning NO₂ in the Arctic

Xin Zhang,* Ronald van der A, Jieying Ding, Henk Eskes, Jos van Geffen, Yan Yin,* Juliëtte Anema, Chris Vagasky, Jeff L. Lapierre, and Xiang Kuang



Cite This: *Environ. Sci. Technol.* 2023, 57, 2322–2332



Read Online

ACCESS |

Metrics & More

Article Recommendations

Supporting Information

ABSTRACT: The Arctic region is experiencing notable warming as well as more lightning. Lightning is the dominant source of upper tropospheric nitrogen oxides (NO_x), which are precursors for ozone and hydroxyl radicals. In this study, we combine the nitrogen dioxide (NO₂) observations from the TROPOspheric Monitoring Instrument (TROPOMI) with Vaisala Global Lightning Dataset 360 to evaluate lightning NO₂ (LNO₂) production in the Arctic. By analyzing consecutive TROPOMI NO₂ observations, we determine the lifetime and production efficiency of LNO₂ during the summers of 2019–2021. Our results show that the LNO₂ production efficiency over the ocean is ~6 times higher than over continental regions. Additionally, we find that a higher LNO₂ production efficiency is often correlated with lower lightning rates. The summertime lightning NO_x emission in the Arctic (north of 70° N) is estimated to be 219 ± 116 Mg of N, which is equal to 5% of anthropogenic NO_x emissions. However, for the span of a few hours, the Arctic LNO₂ density can even be comparable to anthropogenic NO₂ emissions in the region. These new findings suggest that LNO₂ can play an important role in the upper-troposphere/lower-stratosphere atmospheric chemical processes in the Arctic, particularly during the summer.

KEYWORDS: Arctic, lightning, nitrogen dioxide, satellite measurements, TROPOMI



INTRODUCTION

Warming of the climate system is invoking changes in global lightning.^{1–3} Many modeling schemes have been developed over the past 30 years to predict lightning.^{4–9} Most climate models predict more lightning in the mid-latitudes,^{9–12} but predictions for the tropics are currently diverging.^{13,14} In the Arctic, where warming is 4 times faster than the global average, relatively few studies have been conducted.¹⁵ Recently, Chen et al.¹⁶ developed a lightning modeling scheme for the Arctic. Their study suggested a 74–150% increase in the rate of lightning in the permafrost region by the end of the century for a 3.7 °C global mean temperature increase. Ground-based lightning observations support this conclusion, showing that the relative fraction of Arctic lightning over global lightning has tripled from 2010 to 2020.¹⁷ In addition, increased summer moisture convergence in the Arctic is expected to strengthen convective systems, leading to more lightning.¹⁸

Lightning produces nitrogen oxide (NO) through the dissociation of O₂ and N₂ at high temperatures.¹⁹ NO is rapidly oxidized to nitrogen dioxide (NO₂), and an equilibrium is established between NO and NO₂. NO₂ can be detected by satellite, making it a useful tracer for studying lightning processes. Global lightning NO_x (LNO_x) emissions are estimated to be 5 ± 3 Tg of N year⁻¹ and are the dominant natural source of NO_x in the upper troposphere.²⁰ Lightning NO₂ (LNO₂) has a longer lifetime in the upper troposphere

due to the colder temperature and lower reaction rate.^{21,22} Its lifetime ranges from 2 to 12 h near the initial convection and increases to at least 2–3 days downwind of convection,²³ leading to more ozone (O₃) after several days.^{24–26} O₃ is known as a significant greenhouse gas,²⁷ thus, LNO_x has an indirect effect on climate forcing. Furthermore, the photolysis of O₃ produces oxygen atoms in a higher-energy state, which react with water vapor to produce the hydroxyl radical (OH), an atmospheric oxidant. Greater LNO_x emissions shift the equilibrium between OH and HO₂ toward more OH.²⁸ Lightning also directly creates OH through the dissociation of H₂O by visible flashes and subvisible charges.^{29–32} The increase in the level of OH will ultimately reduce the levels of carbon monoxide (CO) and methane (CH₄), another greenhouse gas.^{33,34} Lightning can also trigger wildfires that emit carbon dioxide in the Arctic.¹⁶ Therefore, a better understanding of Arctic LNO_x emissions is important for determining trace gas changes in the Arctic.

Received: October 28, 2022

Revised: January 25, 2023

Accepted: January 25, 2023

Published: February 1, 2023



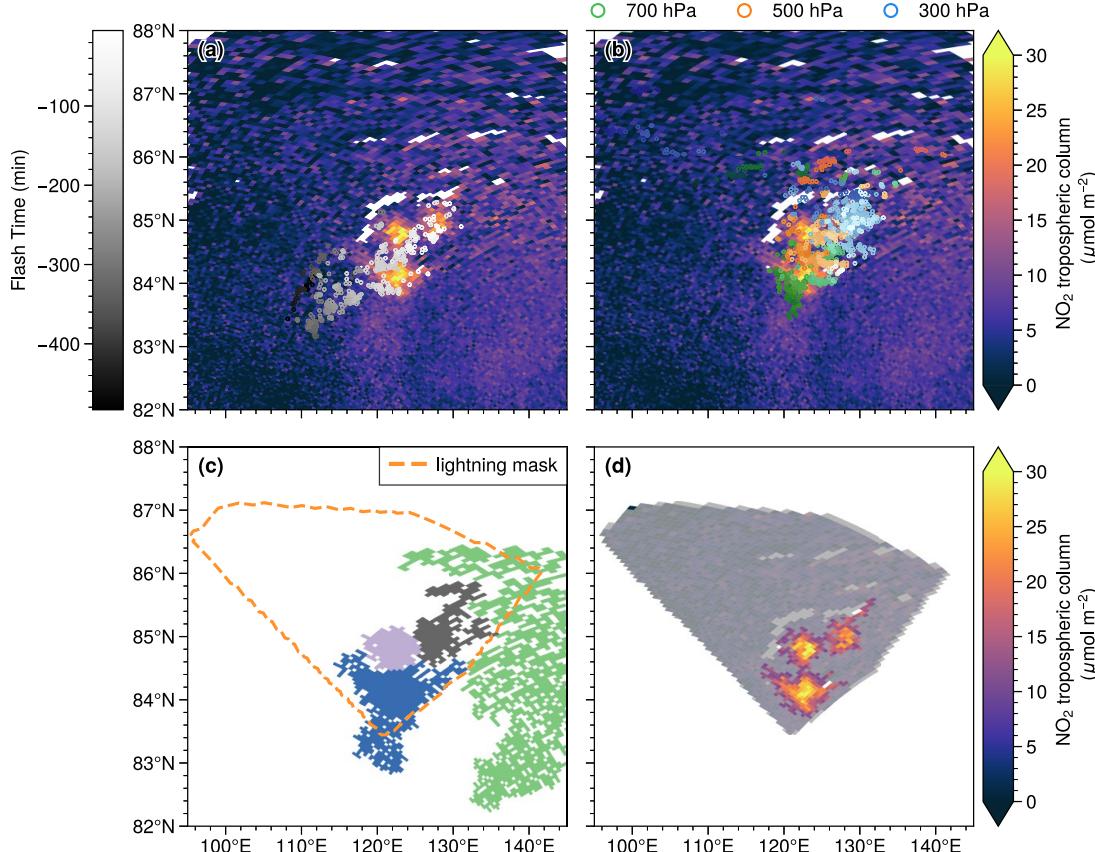


Figure 1. Overview of the process for identifying lightning NO₂ pixels from TROPOMI orbit 09458 on August 1, 2019. The TROPOMI-detected NO₂ tropospheric columns are overlaid with (a) observed lightning strokes and (b) transported air parcels of lightning NO₂ at three pressure levels [300 hPa (blue), 500 hPa (orange), and 700 hPa (green)]. (c) Parcels are combined into one lightning mask (orange circle), which overlaps with high NO₂ selections (filled pixels). The different pixel colors represent different NO₂ selections based on multiple NO₂ thresholds. (d) Final selection (bright colored pixels) of NO₂ tropospheric columns affected by lightning NO₂ within the mask.

Estimating LNO_x requires knowledge of lightning counts and the production efficiency (PE, NO_x per lightning). While there have been several studies on Arctic lightning counts,^{16,17,35} the LNO_x PE for this region is not well quantified. Previous laboratory studies, as well as airborne and satellite-based observations, have estimated that the PE for individual flashes ranges from ≈5 to >1000 mol of NO_x.²⁰ It is uncertain whether mid-latitude lightning is more efficient at producing NO_x than tropical lightning. Some findings from individual storms suggest that this may be the case,^{36–38} but recent satellite-based observations suggest little difference.^{39,40} Considering the regional differences and limited work at the high latitudes, we focus on quantifying the LNO₂ PEs in the Arctic. In this study, we evaluate the NO₂ observations from the TROPOspheric Monitoring Instrument (TROPOMI) with lightning strokes detected by Vaisala Global Lightning Dataset 360 (GLD360). Using consecutive TROPOMI orbits, we are able to determine the LNO₂ lifetime and PEs in the Arctic for the first time, providing insight into regional differences in LNO₂ emissions.

■ DATA AND METHODOLOGY

Lightning Data. The GLD360 network has been in operation since 2009 and consists of very-low-frequency lightning detection sensors located around the world. These sensors can detect both cloud-to-ground (CG) and intracloud (IC) strokes.^{41–43} The detection efficiencies of CG and IC

strokes over the Arctic ocean are estimated to be 50–60% and <10%, respectively.⁴⁴ In the Arctic, the total number of lightning strokes is adjusted to be 4 times the number of detected strokes, on the basis of the constant ratio (≈ 1) of IC to CG strokes in this region. The constant ratio is calculated using two factors: the number of strokes per flash (flash multiplicity) and the mean ratio of IC to CG flashes (also known as the Z ratio) in the high latitudes. The flash multiplicity is highly sensitive to both the detection efficiency and the algorithm used to group strokes into flashes.^{45–48} The mean multiplicity value for high-latitude studies is estimated to be 1.2.⁴⁹ In addition, the mean Z ratio of high-latitude flashes is between 1 and 1.3.^{50–52} To evaluate the sensitivity of the constant ratio, we have used different values (1:2 and 2:1) in Table S4 to assess the uncertainty of estimated LNO_x emissions.

To evaluate the lightning distribution changes at high latitudes, we compare the lightning stroke data from the GLD360 data (2019–2021) with lightning flashes detected by the Optical Transient Detector (OTD, 1996–1999).⁵³ The OTD data product includes only the number of flashes, while one flash can contain one or more strokes. Because the official GLD360 product includes only lightning strokes and our purpose is to investigate changes in lightning distribution, we do not group strokes into flashes. The GLD360 data cover the entire high-latitude region (>60° N), while the OTD instrument observed lightning only south of 75° N. During

the summer season (June–August), the fraction of GLD360 strokes occurring north of 75° N is 3% for the period of 2019–2021 (Figure S2).

NO₂ Satellite Measurements. The TROPOMI instrument is on board the Sentinel 5 Precursor (SSP) satellite, flying in a near-polar, sun-synchronous orbit.⁵⁴ The nadir pixel resolution has been enhanced from 7 × 3.5 to 5.5 × 3.5 km² since August 6, 2019. Its wide swath width (~2600 km) results in overlapping ground pixels for high latitudes. For example, a high-latitude site north of 70° N can have up to seven overpasses in summer, as the detection is also available during the descending orbit. This means that Arctic NO₂ can be tracked from space with an interval of 100 min, the period of a single orbit.

The TROPOMI NO₂ product is the SSP Product Algorithm Laboratory (SSP-PAL) reprocessing product, version 2.3.1, which has improved validity over bright storm scenes compared to earlier versions.⁵⁵ To minimize the impact of boundary layer NO₂ and improve the accuracy of estimating the background, we consider only observations north of 70° N, where the summertime average NO₂ column density is relatively low [$<15 \mu\text{mol m}^{-2}$ (Figure 3a)]. We also exclude pixels with large retrieval errors (processing quality flags >0).

Estimation of Lightning NO₂. The LNO₂ estimation in this study involves three main steps: (1) identifying high NO₂ columns associated with lightning strokes using wind data, (2) calculating LNO₂ columns using a new algorithm, and (3) determining the LNO₂ production efficiency using consecutive TROPOMI observations.

Procedure for Lightning Clustering. We cluster lightning strokes within a 40 km radius of each stroke and a 12 h period⁵⁶ before the TROPOMI overpass time using the Density-Based Spatial Clustering of Applications with Noise (DBSCAN) algorithm.^{57,58} Because wildfires can also produce NO₂, the lightning clusters affected by wildfire emissions are identified and filtered out using the Visible/Infrared Imager/Radiometer Suite (VIIRS) 375m active fire product. The VIIRS is on board the Suomi National Polar-orbiting Partnership (Suomi-NPP), which is in the same orbit but 3.5 min ahead of SSP. We refer to lightning clusters without fires inside as “clean” clusters.

For each clean lightning cluster, we define air parcels affected by LNO₂ at three pressure levels (300, 500, and 700 hPa) on the basis of the time and location of detected strokes (Figure 1a). The air parcels containing LNO₂ are then transported by horizontal advection (Figure 1b). To determine the location of air parcels at the TROPOMI overpass time, we

use the hourly fifth generation of atmospheric reanalysis (ERAS) wind data.⁵⁹ The isobaric forward trajectories at the three pressure levels are used to construct an approximate LNO₂ region. The final locations of air parcels are combined into one lightning mask (the orange circle in Figure 1c), which is used to determine the background NO₂ and LNO₂ pixels.

In this study, we use a watershedding technique to derive the area of high NO₂ vertical column density (V_{NO_2} , pixels in Figure 1c). This method treats the pixel values as a topographic surface and separates them into individual regions known as catchment basins.^{60,61} Threshold values ranging from 4×10^{14} to 1×10^{15} molecules cm⁻² are applied in steps of 2×10^{14} molecules cm⁻² to detect multiple localized high-NO₂ features. These features are used to identify nearby areas with high V_{NO_2} values (pixels of different colors in Figure 1c). Detailed steps of the watershedding process are described by Heikenfeld et al.⁶¹ The lightning cluster containing one or more high- V_{NO_2} areas is selected for further analysis of LNO₂. The final LNO₂ area (bright colored pixels in Figure 1d) is the region with a high V_{NO_2} that is reidentified by the watershedding method within the lightning mask (see the Supporting Information for details).

Calculation of the Lightning NO₂ Vertical Column. As discussed by Zhang et al.,²⁸ the official TROPOMI algorithm does not include the LNO₂ in the NO₂ retrieval. Thus, we subtract the background NO₂ from the official S_{NO_2} and convert the difference to the tropospheric LNO₂ vertical column with an air mass factor (AMF). The algorithm is expressed as

$$V_{\text{LNO}_2} = \frac{S_{\text{NO}_2} - S_{\text{BG}}}{\text{AMF}_{\text{LNO}_2}} \quad (1)$$

where V_{LNO_2} is the tropospheric LNO₂ vertical column, S_{NO_2} is the TROPOMI-measured tropospheric NO₂ slant column, and $\text{AMF}_{\text{LNO}_2}$ is a customized lightning air mass factor. Following Allen et al.,⁵⁶ the background S_{NO_2} (S_{BG}) is defined as the product of the tropospheric AMF and the 30th percentile of V_{NO_2} over low-NO₂ pixels within the lightning mask. The low-NO₂ region (gray pixels in Figure 1d) includes all masked pixels outside of the high- V_{NO_2} regions identified by the watershedding technique. $\text{AMF}_{\text{LNO}_2}$ is the ratio of the “visible” LNO₂ slant column to the total tropospheric LNO₂ vertical column:

$$\text{AMF}_{\text{LNO}_2} = \frac{(1 - f_r) \int_{p_{\text{surf}}}^{p_{\text{tp}}} w_{\text{clear}}(p) \text{LNO}_2(p) dp + f_r \int_{p_{\text{cloud}}}^{p_{\text{tp}}} w_{\text{cloudy}}(p) \text{LNO}_2(p) dp}{\int_{p_{\text{surf}}}^{p_{\text{tp}}} \text{LNO}_2(p) dp} \quad (2)$$

where f_r is the cloud radiance fraction in the NO₂ window, p_{surf} is the surface pressure, p_{cloud} is the cloud pressure, p_{tp} is the tropopause pressure, w_{clear} and w_{cloudy} are the pressure-dependent scattering weights from the lookup table⁶² for clear and cloudy parts, respectively, and $\text{LNO}_2(p)$ is a pressure-dependent tropospheric NO₂ profile presented by a modified Gaussian distribution. The peak width of the Gaussian distribution is set to 180 hPa following Ott et al.⁶³ and Luo et al.⁶⁴ (see the Supporting Information for details), and the

peak level is the highest TROPOMI cloud pressure in the lightning mask.

The scattering weights used in the calculation of $\text{AMF}_{\text{LNO}_2}$ are determined by five parameters: surface pressure, solar zenith angle, viewing zenith angle, relative azimuth angle, and albedo. For the cloudy part, the surface pressure and albedo values used in the lookup table are specific to the clouds present. The surface pressure is set equal to the cloud pressure, and the albedo is set equal to the cloud albedo. The cloud

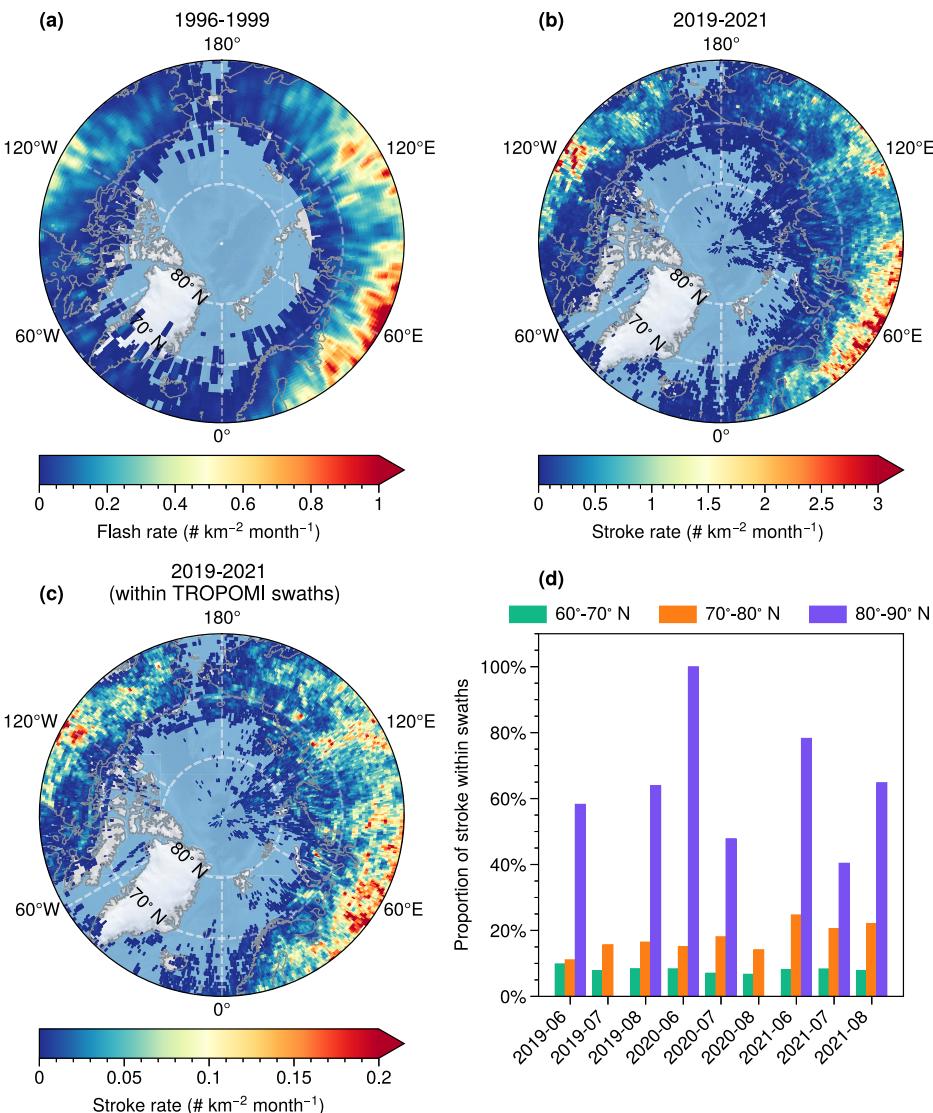


Figure 2. (a) Mean OTD lightning flash rate in June–August 1996–1999. (b) Mean GLD360 lightning stroke rate of June–August 2019–2021. (c) Same as panel b but counting only the lightning inside the TROPOMI swaths during the 3 h period before the TROPOMI overpass time. Grids with no lightning appear as the light-blue backgrounds in panels a–c. (d) Monthly ratio of panel c to panel b.

pressure detected by TROPOMI represents a pressure level inside the cloud rather than the geometrical cloud top.^{65,66} It ranges from 130 to 987 hPa, with the most frequent values in the 250–300 hPa bin (Figure S3). The assumed LNO₂ profile should be more realistic than the default a priori NO₂ profile, because the high NO₂ is inside the cloud and the above-cloud NO₂ is low due to photolysis.⁶⁷ For high and thick clouds, the scattering weights are fairly uniform above the clouds, and the sensitivity of LNO₂ is highest around the middle of cloud, which is close to the cloud pressure detected by TROPOMI.⁶⁷ This minimizes the effects of possibly wrongly assumed NO₂ peak pressures on the retrieval.⁶⁸ Note that we find some cases with clouds higher than the tropopause. We dismiss these cases because the NO₂ concentration could be affected by stratospheric O₃.^{28,69}

Estimation of the Lightning NO₂ Production Efficiency. As mentioned in **NO₂ Satellite Measurements**, TROPOMI overpasses the same site north of 70° N several times. Thus, two consecutive orbits can observe the same thunderstorm. To make it easier to analyze patterns and variations in Arctic LNO₂ cases, we have created an interactive Web site (<https://arctic-lightning-no2.streamlit.app/>) that allows users to filter cases and investigate the relationship among LNO₂, cloud pressure, and lightning interactively.

The relationship between V_{LNO_2} (moles per square meter) at two timestamps can be defined as

$$\sum_{P(T_2)} V_{\text{LNO}_2 i} A_i = e^{-(T_2 - T_1)/\tau} \sum_{P(T_1)} V_{\text{LNO}_2 j} A_j + \text{PE} \sum_N e^{-(T_2 - t_k)/\tau} \quad (3)$$

where p is the number of pixels in the LNO₂ area, A is the area (square meters) of each pixel, T is the TROPOMI overpass time, t is the lightning occurrence time, τ is the near-convection LNO₂ lifetime, PE is the LNO₂ production efficiency (moles per stroke), and N is the total number of strokes during the interval between consecutive orbits. Specifically, the exponential component considers the chemical loss of NO₂. In eq 3, i and j are the pixel indices for the TROPOMI instrument, A_i and A_j are different LNO₂ areas taking advection into account (Figure S4), T_1 and T_2 represent the mean overpass times of the TROPOMI instrument over

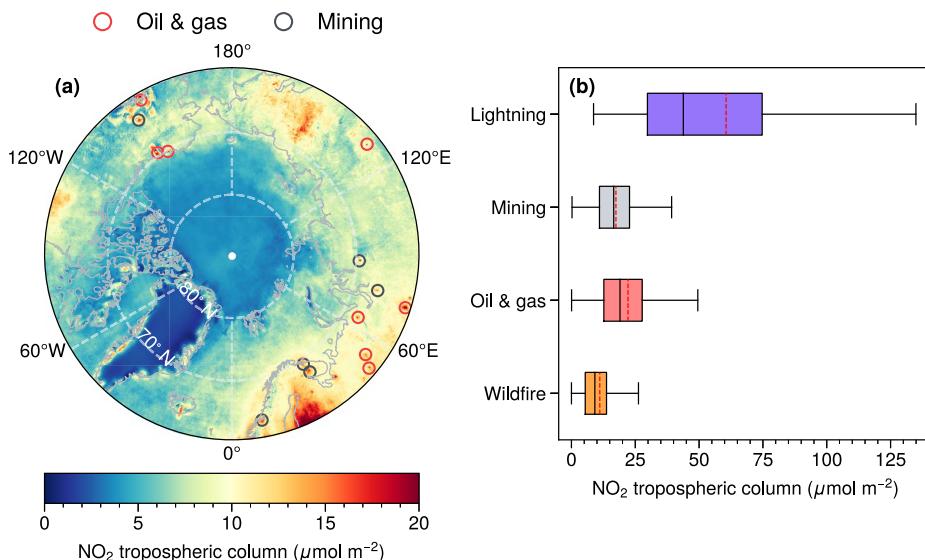


Figure 3. (a) Mean 4 km × 4 km TROPOMI tropospheric NO₂ column density in the local afternoon during June–August 2019–2021. The mining and oil and gas stations are shown as gray and red circles, respectively. (b) Comparisons of NO₂ among four sources: lightning, mining, oil and gas, and wildfire. The lightning bar represents the maximum NO₂ values over pixels for each lightning case, while the wildfire, mining, and oil and gas bars are the daily maximum NO₂ values at typical locations. The box plots indicate the median (black line) and mean (red line) values; the lower and upper boundaries represent the 25th and 75th percentiles, respectively, and the lower and upper error lines are the 10th and 90th percentiles, respectively.

the LNO₂ region for two consecutive orbits, and t_k represents the time of the k th lightning stroke (where k ranges from 1 to N) that occurs between the two orbits.

Note that if there is no lightning between two consecutive orbits, eq 3 can be simplified as

$$\sum_{p(t_2)} V_{\text{LNO}_2} A_i = e^{-(T_2 - T_1)/\tau} \sum_{p(T_1)} V_{\text{LNO}_2} A_j \quad (4)$$

We calculate the value of τ as 3 ± 1 h by analyzing the data from specific cases with high LNO₂ but no lightning between orbits (Table S2 and Figure S4). We then substitute the 3 h τ in eq 3 to calculate the LNO₂ PEs for the remaining lightning cases.

Previous studies have implemented three other methods to estimate LNO₂ production in tropical and mid-latitude regions, including (1) using linear regression to correlate background NO₂ plus LNO₂ (LNO₂*) with lightning,^{39,70,71} (2) deriving LNO₂ directly using customized air mass factors,^{28,67,72} and (3) deriving LNO₂* and subtracting background NO₂ derived from aircraft observations^{70,73} or average NO₂ over non-lightning pixels.^{38,40,56} However, the linear regression method is not suitable for our study due to variations in background NO₂ with solar zenith angle. The second approach requires detailed LNO₂ simulation with lightning parametrization, which is still quite uncertain,^{13,14,16} particularly in the Arctic where lightning data are limited.¹⁷ Additionally, obtaining mean background NO₂ data sets for each grid cell (i.e., 1° × 1°) in the Arctic is difficult, as TROPOMI detects only a few nonlightning convection cases north of 70° N (not shown).

RESULTS

Lightning Distributions. Figure 2 compares the spatial distribution of summer (June–August) lightning at high latitudes using satellite and ground-based lightning observations. Both data sets show higher rates over the Siberia and Alaska permafrost [60–65° N (Figure 2a,b)], which is the

main Arctic fire regime.⁷⁴ The average OTD lightning flash rate and GLD360 stroke rate are 0.22 and 0.61 km⁻² month⁻¹, respectively. However, OTD records less lightning near the Chukchi Sea and more over the Irminger Sea, which is likely due to interannual variations in Arctic precipitation linked to poleward moisture transport.¹⁸

To determine if LNO₂ can be detected by TROPOMI, we count the number of GLD360 lightning strokes within TROPOMI swaths during a 3 h period [i.e., the near-convection LNO₂ lifetime (see Estimation of the Lightning NO₂ Production Efficiency)] before each overpass. TROPOMI does not continuously measure the Arctic, but it covers large parts of the region in summer with 14 overlapping orbits per day. The number of within-swath lightning strokes (Figure 2c) is ~9% of the total counts and follows the same geographical pattern as the total strokes (Figure 2b). The proportion of within-swath lightning strokes increases with latitude from <10% (60–70° N) to 10–25% (70–80° N) and 40–100% [80–90° N (Figure 2d)] due to increased overlap of swaths at higher latitudes. Thus, the TROPOMI NO₂ observations are powerful for analyzing LNO₂ in the Arctic, where ground-based⁷⁵ and aircraft-based⁷⁶ NO₂ observations are limited. We choose to focus on the region north of 70° N for LNO₂ analysis due to the greater TROPOMI coverage and fewer NO₂ emissions from other sources (e.g., wildfires or gas production). The number of summer strokes detected by GLD360 is 1.2×10^6 (2019), 1.6×10^6 (2020), and 9.8×10^5 (2021) north of 70° N (Table S1). In the region with the highest TROPOMI coverage (80–90° N), the number of strokes in 2021 is $\sim 2.9 \times 10^4$, nearly twice as many as the total number recorded over the previous nine years.⁷⁷ The northernmost lightning stroke reaches 89.5° N, which was also reported (89.6° N) by the World Wide Lightning Location Network (WWLLN).¹⁷

Anthropogenic versus Lightning NO₂. Figure 3a shows the mean NO₂ column densities as measured by TROPOMI at high latitudes from June to August 2019–2021. While the

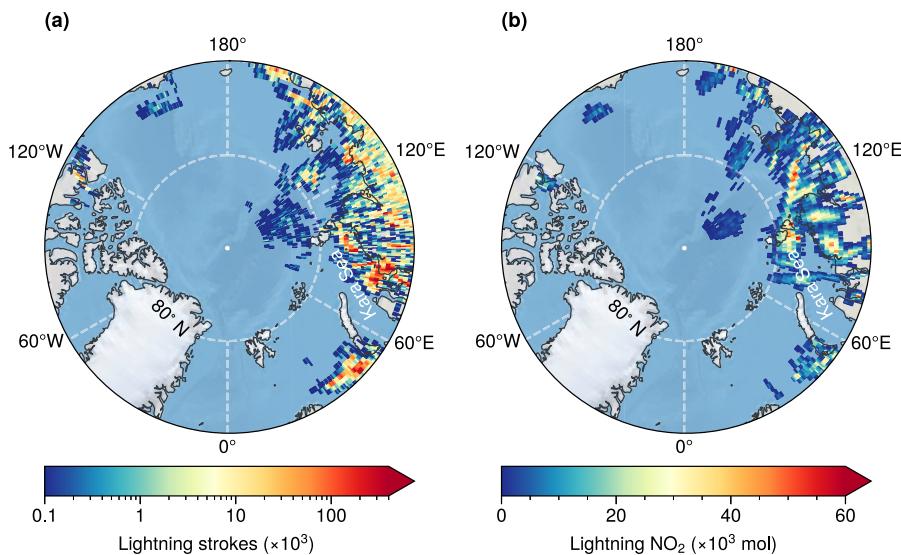


Figure 4. (a) Sum of GLD360 lightning strokes for selected cases per $0.5^\circ \times 0.5^\circ$ grid. (b) Sum of lightning NO₂ derived from TROPOMI observations for selected cases per $0.5^\circ \times 0.5^\circ$ grid.

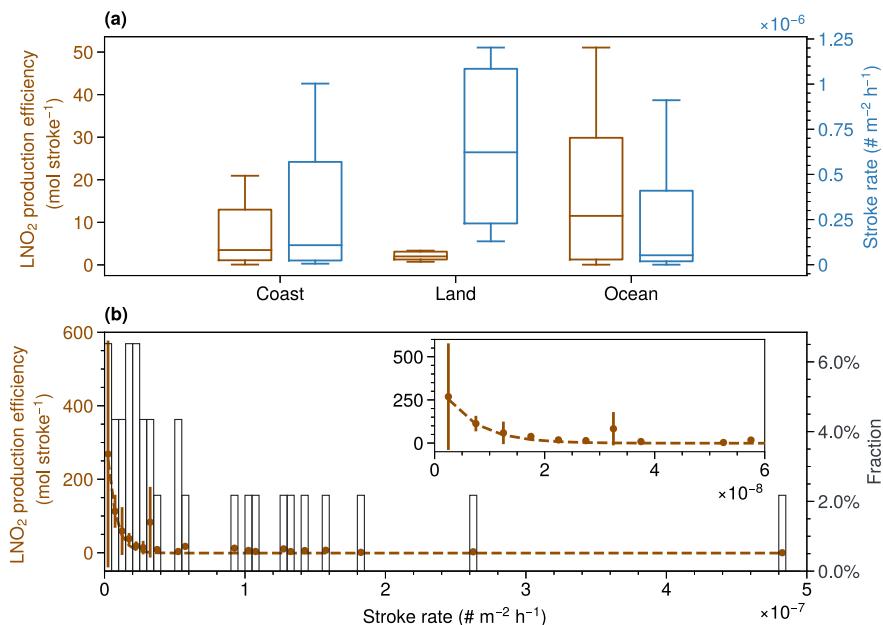


Figure 5. (a) Comparison of lightning NO₂ (LNO₂) production efficiency (brown) and stroke rate (blue) for three regions: coast, land, and ocean. The coast region is defined as a 500 m radius around the coastline. The box plots indicate the median values; the lower and upper boundaries represent the 25th and 75th percentiles, respectively, and the lower and upper error lines are the 10th and 90th percentiles, respectively. (b) LNO₂ production efficiency vs stroke rate, with histograms of case fractions in each $5 \times 10^{-9} \text{ m}^{-2} \text{ h}^{-1}$ stroke rate bin overlaid.

averaged LNO₂ disappears into the background, NO₂ enhancements can still be observed over urban, industrial, and wildfire regions. For example, NO₂ correlated with mining operations ($17 \pm 2 \mu\text{mol m}^{-2}$) in Alaska, Norway, and Russia is clearly visible. Other sources of NO₂ include oil and gas activities associated with the Mackenzie Valley in Arctic Canada, Prudhoe Bay/Kuparuk in North Alaska, and Yamal gas pipeline/Urengoy gas fields in Russia.⁷⁸ The NO₂ spikes along the Greenland coast may be false signals caused by retrieval complications due to the complicated topography of the region.⁷⁹ Contrary to NO₂ pollution, TROPOMI observations north of 70° N show low background NO₂ ($4.4 \pm 1.3 \mu\text{mol m}^{-2}$), which is 45% lower than the mean NO₂ between 60° N and 65° N.

By comparing the highest LNO₂ column ($61 \pm 50 \mu\text{mol m}^{-2}$) of all cases with typical anthropogenic and wildfire NO₂ at high latitudes (Figure 3b), we find that the median LNO₂ column density is triple, although the time scale of emission is on the order of hours. One particularly notable event is the deep convection occurring over the Laptev Sea (Figure S5), where the maximum LNO₂ is $246 \mu\text{mol m}^{-2}$, comparable to the highest NO₂ column density ($234 \mu\text{mol m}^{-2}$) in the United States.⁸⁰ Given the significant contribution of LNO₂ for a few hours, it is important to calculate LNO₂ emissions in the Arctic.

Lightning NO₂ Production Efficiency and Emission. LNO₂ emission is the product of stroke counts and LNO₂ PEs (LNO₂ per stroke). It is impossible to derive the LNO₂ PE

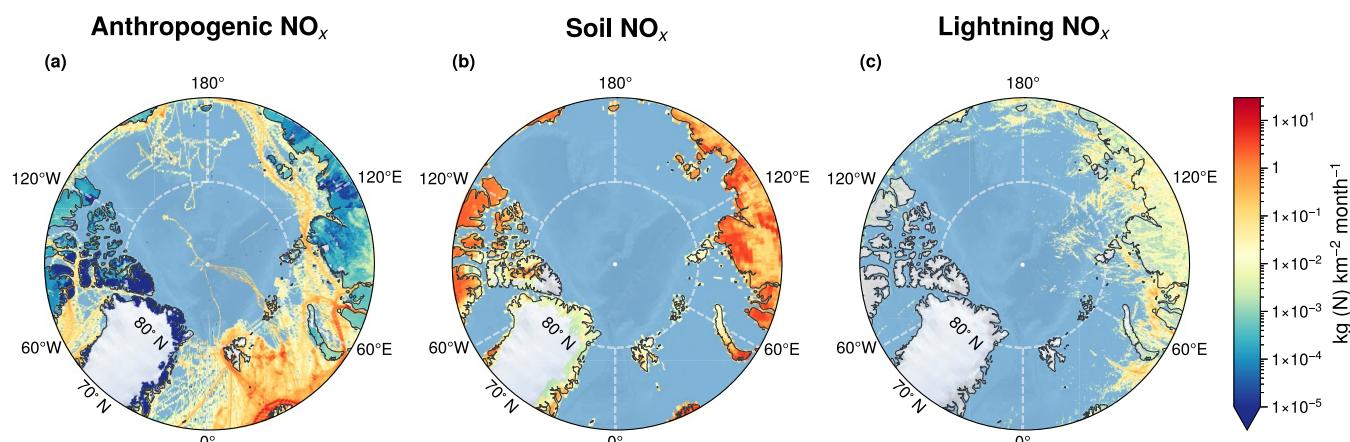


Figure 6. Monthly NO_x emissions in the Arctic from June to August. (a) Anthropogenic emissions including ship emissions, (b) soil emissions, and (c) lightning emissions. The lightning NO_x emissions are the mean values from 2019 to 2021, while the other emissions are from the 2018 Copernicus Atmosphere Monitoring Service (CAMS) global emission inventories.

directly on the basis of data from a single orbit, because the LNO₂ detected by TROPOMI includes both production and depletion, and a second orbit is needed for a time dimension. Here we utilize the LNO₂ from consecutive orbits to estimate the LNO₂ PEs (see [Estimation of the Lightning NO₂ Production Efficiency](#) for details). On the basis of the screening of LNO₂ and lightning strokes, we choose 43 case studies with a total of 115 orbits. Cases between 60° W and 180° W are excluded due to insufficient LNO₂ or a lack of observations. Most cases are located between 60° E and 180° E ([Figure 4a](#)). By comparing the lightning distribution ([Figure 4a](#)) with the summed LNO₂ within a 0.5° × 0.5° grid box ([Figure 4b](#)), we find that the LNO₂ observed by TROPOMI has been transported away from the lightning source. For instance, although there is no lightning over the Kara Sea (77–80° N, 65–105° E), the LNO₂ in the region is $\sim 5.1 \times 10^6$ mol in total. This arises from the upper tropospheric wind transporting LNO₂ in the dominant downwind direction toward the east, highlighting the importance of matching lightning and NO₂.

Despite the transport of LNO₂, the comparison between the distribution of lightning and LNO₂ reveals a strong spatial correlation between the two phenomena. As a result, we estimate the LNO₂ PE for each case study. The median LNO₂ PEs are 2.0 (25th to 75th percentile, 1.3–3.1) mol stroke⁻¹, 3.5 (25th to 75th percentile, 1.1–13.0) mol stroke⁻¹, and 11.5 (25th to 75th percentile, 1.3–29.9) mol stroke⁻¹ over land, coast, and ocean, respectively ([Figure 5a](#)). In addition to these median values, we also measure the top 10 highest LNO₂ PEs, which range from 27 to 612 mol stroke⁻¹ ([Table S3](#)). These highest values are all found in oceanic or coastal areas. Due to the limited number of cases, it is not possible to determine if there is a statistically significant difference in LNO₂ PEs between ocean and land. However, the 2-fold larger LNO₂ PE in marine locations than in continents has also been found in the tropics and mid-latitudes^{39,40,81} due to the higher flash energy of ocean-based lightning.^{82,83}

Recent studies suggest that LNO₂ PE is related to flash size, with larger flashes producing more LNO₂.^{36,81} Furthermore, stronger updrafts are associated with smaller flashes and higher lightning rates.^{84–86} Therefore, we calculate the stroke rate, which is the number of lightning strokes per unit LNO₂ area per unit time. It ranges from 2.3×10^{-10} to 4.1×10^{-6} m⁻² h⁻¹,

with the continental stroke rate (6.2×10^{-7} m⁻² h⁻¹) being ~ 11 times higher than over the ocean (5.2×10^{-8} m⁻² h⁻¹). We find an approximate power law relationship between the stroke rate and LNO₂ PE ([Figure 5b](#)). When the stroke rate decreases by 2 orders of magnitude, LNO₂ PE increases by a factor of 10, which is consistent with mid-latitude studies.^{40,72}

Because the area applied in the stroke rate calculation depends on the LNO₂ selection from TROPOMI, we multiply the number of lightning strokes in each of the three regions by the median PEs of these regions and sum the results to obtain the total Arctic LNO₂ emissions [$>70^\circ$ N ([Table S1](#))]. In 2020, LNO₂ emission is 109 Mg of N, which is 31% higher than the mean LNO₂ emission of 83 Mg of N in 2019 and 2021. The dominant contributor to this increase is the enhanced emission (35%) between 70° N and 80° N, where the mean convective available potential energy (CAPE) is 32% higher in 2020 ([Table S1](#) and [Figure S8](#)). Note that LNO₂ emission (4.3 Mg of N) near the pole (80–90° N) increases by 353% in 2021 due to an increase in lightning activity ([Table S1](#)). Overall, we estimate the summer LNO₂ emission in the Arctic as 30 ± 16 Mg of N month⁻¹ considering the uncertainties [53% ([Table S4](#))].

Assuming the ratio of NO_x to NO₂ as 2.4,⁸⁷ we estimate the LNO_x emission to be 73 Mg of N month⁻¹ and compare it with the anthropogenic and soil NO_x emissions in the Arctic during the summer ([Figure 6](#)). We find that soil emissions, totaling 2670 Mg of N month⁻¹, dominate NO_x over the Arctic land, while anthropogenic NO_x emissions of 350 Mg of N month⁻¹ over land are roughly the same in magnitude as wildfire emissions of 430 Mg of N month⁻¹. Ship emissions, totaling 1160 Mg of N month⁻¹, are the primary source of NO_x over the Arctic ocean. However, the LNO_x contributes 93% of NO_x in the northeastern region of the Arctic ocean (90–180° E, 80–90° N). It is important to note that the lifetime of upper tropospheric NO₂ away from thunderstorms is ~ 0.5 –8 days due to low temperatures;^{20,23} therefore, further studies are needed to evaluate how LNO₂ affects O₃, CO, and CH₄ in the Arctic.

DISCUSSION

In this study, we estimate both the LNO₂ lifetime and the production efficiency in the Arctic based on consecutive TROPOMI NO₂ observations. Our results indicate that the 3

h LNO₂ lifetime is similar to that previously reported in the United States.²³ The continental LNO₂ production efficiency [2.0 (1.3–3.1) mol stroke⁻¹] is also comparable to that observed over the United States (1.6 ± 0.1 mol stroke⁻¹).⁷¹ Additionally, our findings suggest that oceanic lightning in the Arctic can produce 6 times higher NO_x than continental lightning. As a result, when the amount of lightning increases by the same amount over the Arctic ocean and land, the oceanic lightning will produce more LNO₂. Overall, the mean LNO_x emissions (219 Mg of N) in summer (June–August) are equal to 5% of anthropogenic NO_x emissions in the Arctic.

One limitation of our study is that we derive the LNO₂ column density without retrieving LNO₂ vertical profiles. To obtain these profiles, techniques such as cloud slicing can be used,^{88,89} but they require a large sample size to reduce noise. Future aircraft observations, like the Deep Convective Clouds and Chemistry (DC3) project,⁹⁰ could provide more detailed LNO₂ profiles and improve our understanding of LNO_x and air pollution in the Arctic.^{75,91}

Accurate observations, simulations, and validations of Arctic lightning are crucial for understanding its impact on LNO₂ and other atmospheric processes, as global warming is expected to lead to an increase in its occurrence. However, the detection efficiency of ground-based lightning networks in the Arctic is still low due to the large areas of ocean and ice coverage in the region.⁴⁴ Additionally, current parametrizations of Arctic lightning tend to focus on land-based processes, particularly permafrost.¹⁶ To address these challenges, consistent satellite observations, such as those provided by OTD, are essential. Note that the Lightning Imaging Sensors (LIS) on board the Tropical Rainfall Measuring Mission (TRMM)⁹² and the International Space Station (ISS)⁹³ are primarily designed to detect tropical and mid-latitude lightning, with TRMM LIS covering low latitudes ($\pm 38^\circ$) and ISS LIS extending coverage to higher latitudes (up to $\pm 55^\circ$). The range of latitudes that can be detected by future planned lightning sensors is currently unknown. However, carrying these sensors, such as the Geostationary Lightning Mapper (GLM) or the Lightning Mapping Instrument (LMI),^{94,95} on hydrometeorological satellites like Arktika-M⁹⁶ could greatly benefit Arctic lightning studies by providing more comprehensive coverage of this region.

■ ASSOCIATED CONTENT

Data Availability Statement

The retrieval algorithm is available at <https://github.com/zxdawn/SSP-LNO2>.⁹⁷ The relevant data and analysis code are hosted at <https://github.com/zxdawn/SSP-LNO2-Notebook>.^{98,99} We also established a Web site (<https://arctic-lightning-no2.streamlit.app/>) to provide an interactive interface for exploring Arctic lightning NO₂ cases. The data supporting the findings of this study are openly available: SSP-PAL TROPOMI data (<https://data-portal.ssp-pal.com>), NASA FIRMS VIIRS active fire product (<https://earthdata.nasa.gov/firms>), LIS/OTD 0.5 Degree High Resolution Monthly Climatology data set (10.5067/LIS/LIS-OTD/DATA303), ERA5 reanalysis data (10.24381/cds.bd0915c6), CAMS global emission inventories (<https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-emission-inventories>), and global biomass burning emissions based on fire radiative power (GFAS, <https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-fire-emissions-gfas>). The VAISALA GLD360 data are available upon request

from VAISALA (<https://www.vaisala.com/en/products/systems/lightning/gld360>).

■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.2c07988>.

Description of a priori lightning LNO₂ profiles; steps of selecting LNO₂ pixels; time series of GLD360 lightning strokes (Figure S2); histogram of TROPOMI cloud pressure and tropopause pressure (Figure S3); schematic overview of LNO₂ selections (Figure S4); large LNO₂ case (Figure S5); relationship between TROPOMI cloud pressure and above-cloud NO₂ column (Figure S6); time series of the flash rate recorded by the Optical Transient Detector (Figure S7); mean GLD360 lightning stroke rate, convective available potential energy, and aerosol optical depth (Figure S8); and tables describing the GLD360 stroke counts, LNO₂ lifetimes, LNO₂ production efficiencies, and uncertainties of LNO₂ estimations ([PDF](#))

■ AUTHOR INFORMATION

Corresponding Authors

Xin Zhang – KNMI-NUIST Center for Atmospheric Composition, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China; Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands; Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China;  orcid.org/0000-0002-1756-6620; Email: xinzhang1215@gmail.com

Yan Yin – KNMI-NUIST Center for Atmospheric Composition, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China; Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China; Email: yinyan@nuist.edu.cn

Authors

Ronald van der A – KNMI-NUIST Center for Atmospheric Composition, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China; Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands

Jieying Ding – Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands;  orcid.org/0000-0003-1263-2876

Henk Eskes – Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands

Jos van Geffen – Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands;  orcid.org/0000-0003-2121-4553

Juliette Anema – Department of Satellite Observations, Royal Netherlands Meteorological Institute (KNMI), 3731 GA De Bilt, The Netherlands; Wageningen University and Research, Meteorology and Air Quality, 6708 PB Wageningen, The Netherlands

Chris Vagasky – Vaisala Inc., Louisville, Colorado 80027, United States

Jeff L. Lapierre – *Earth Networks, Germantown, Maryland 20876, United States*
Xiang Kuang – *Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science and Technology (NUIST), Nanjing 210044, China*

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acs.est.2c07988>

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was funded by the National Natural Science Foundation of China (Grants 91644224 and 42075176) and the China Scholarship Council (202009040001).

REFERENCES

- (1) Reeve, N.; Toumi, R. Lightning Activity as an Indicator of Climate Change. *Q. J. R. Meteorol. Soc.* **1999**, *125*, 893–903.
- (2) Williams, E. R. Lightning and Climate: A Review. *Atmos. Res.* **2005**, *76*, 272–287.
- (3) Price, C. Will a Drier Climate Result in More Lightning? *Atmos. Res.* **2009**, *91*, 479–484.
- (4) Price, C.; Rind, D. A Simple Lightning Parameterization for Calculating Global Lightning Distributions. *J. Geophys. Res. Atmos.* **1992**, *97*, 9919–9933.
- (5) Price, C.; Penner, J.; Prather, M. NO_x from Lightning: 1. Global Distribution Based on Lightning Physics. *J. Geophys. Res. Atmos.* **1997**, *102*, 5929–5941.
- (6) Allen, D. J.; Pickering, K. E. Evaluation of Lightning Flash Rate Parameterizations for Use in a Global Chemical Transport Model. *J. Geophys. Res.: Atmos.* **2002**, *107*, ACH 15-1–ACH 15-21.
- (7) Futyan, J. M.; Del Genio, A. D. Relationships between Lightning and Properties of Convective Cloud Clusters. *Geophys. Res. Lett.* **2007**, *34*, L15705.
- (8) Finney, D. L.; Doherty, R. M.; Wild, O.; Huntrieser, H.; Pumphrey, H. C.; Blyth, A. M. Using Cloud Ice Flux to Parametrise Large-Scale Lightning. *Atmos. Chem. Phys.* **2014**, *14*, 12665–12682.
- (9) Romps, D. M.; Seeley, J. T.; Vollaro, Molinari, J. Projected Increase in Lightning Strikes in the United States Due to Global Warming. *Science* **2014**, *346*, 851–854.
- (10) Price, C.; Rind, D. Modeling Global Lightning Distributions in a General Circulation Model. *Mon. Wea. Rev.* **1994**, *122*, 1930–1939.
- (11) Michalon, N.; Nassif, A.; Saouri, T.; Royer, J. F.; Pontikis, C. A. Contribution to the Climatological Study of Lightning. *Geophys. Res. Lett.* **1999**, *26*, 3097–3100.
- (12) Luhar, A. K.; Galbally, I. E.; Woodhouse, M. T.; Abraham, N. L. Assessing and Improving Cloud-Height-Based Parameterisations of Global Lightning Flash Rate, and Their Impact on Lightning-Produced NO_x and Tropospheric Composition in a Chemistry-Climate Model. *Atmos. Chem. Phys.* **2021**, *21*, 7053–7082.
- (13) Finney, D. L.; Doherty, R. M.; Wild, O.; Stevenson, D. S.; MacKenzie, I. A.; Blyth, A. M. A Projected Decrease in Lightning under Climate Change. *Nat. Clim. Chang.* **2018**, *8*, 210–213.
- (14) Romps, D. M. Evaluating the Future of Lightning in Cloud-resolving Models. *Geophys. Res. Lett.* **2019**, *46*, 14863–14871.
- (15) Rantanen, M.; Karpechko, A. Y.; Lippinen, A.; Nordling, K.; Hyvärinen, O.; Ruosteenoja, K.; Vihma, T.; Laaksonen, A. The Arctic Has Warmed Nearly Four Times Faster than the Globe since 1979. *Communications Earth & Environment* **2022**, *3*, 1–10.
- (16) Chen, Y.; Romps, D. M.; Seeley, J. T.; Veraverbeke, S.; Riley, W. J.; Mekonnen, Z. A.; Randerson, J. T. Future Increases in Arctic Lightning and Fire Risk for Permafrost Carbon. *Nat. Clim. Chang.* **2021**, *11*, 404–410.
- (17) Holzworth, R. H.; Brundell, J. B.; McCarthy, M. P.; Jacobson, A. R.; Rodger, C. J.; Anderson, T. S. Lightning in the Arctic. *Geophys. Res. Lett.* **2021**, *48*, e2020GL091366.
- (18) Bintanja, R.; van der Wiel, K.; van der Linden, E. C.; Reusen, J.; Bogerd, L.; Krikken, F.; Selten, F. M. Strong Future Increases in Arctic Precipitation Variability Linked to Poleward Moisture Transport. *Sci. Adv.* **2020**, *6*, eaax6869.
- (19) Zel'dovich, Y.; Raizer, Y. In *Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena*; Hayes, W. D., Probstein, R. F., Zel'dovich, Y., Raizer, Y., Eds.; Academic Press, 1967; pp 566–571.
- (20) Schumann, U.; Huntrieser, H. The Global Lightning-Induced Nitrogen Oxides Source. *Atmos. Chem. Phys.* **2007**, *7*, 3823–3907.
- (21) Jaeglé, L.; Jacob, D. J.; Wang, Y.; Weinheimer, A. J.; Ridley, B. A.; Campos, T. L.; Sachse, G. W.; Hagen, D. E. Sources and Chemistry of NO_x in the Upper Troposphere over the United States. *Geophys. Res. Lett.* **1998**, *25*, 1705–1708.
- (22) Martin, R. V.; Sauvage, B.; Folkins, I.; Sioris, C. E.; Boone, C.; Bernath, P.; Ziemke, J. Space-Based Constraints on the Production of Nitric Oxide by Lightning. *J. Geophys. Res.: Atmos.* **2007**, *112*, 1479.
- (23) Nault, B. A.; Laughner, J. L.; Wooldridge, P. J.; Crounse, J. D.; Dibb, J.; Diskin, G.; Peischl, J.; Podolske, J. R.; Pollack, I. B.; Ryerson, T. B.; Scheuer, E.; Wennberg, P. O.; Cohen, R. C. Lightning NO_x Emissions: Reconciling Measured and Modeled Estimates With Updated NO_x Chemistry. *Geophys. Res. Lett.* **2017**, *44*, 9479–9488.
- (24) Pickering, K. E.; Thompson, A. M.; Tao, W.-K.; Kucsera, T. L. Upper Tropospheric Ozone Production Following Mesoscale Convection during STEP/EMEX. *J. Geophys. Res. Atmos.* **1993**, *98*, 8737–8749.
- (25) Pickering, K. E.; Thompson, A. M.; Wang, Y.; Tao, W.-K.; McNamara, D. P.; Kirchhoff, V. W. J. H.; Heikes, B. G.; Sachse, G. W.; Bradshaw, J. D.; Gregory, G. L.; Blake, D. R. Convective Transport of Biomass Burning Emissions over Brazil during TRACE A. *J. Geophys. Res. Atmos.* **1996**, *101*, 23993–24012.
- (26) Kang, D.; Mathur, R.; Pouliot, G. A.; Gilliam, R. C.; Wong, D. C. Significant Ground-Level Ozone Attributed to Lightning-Induced Nitrogen Oxides during Summertime over the Mountain West States. *npj Clim. Atmos. Sci.* **2020**, *3*, 6.
- (27) Iglesias-Suarez, F.; Kinnison, D. E.; Rap, A.; Maycock, A. C.; Wild, O.; Young, P. J. Key Drivers of Ozone Change and Its Radiative Forcing over the 21st Century. *Atmos. Chem. Phys.* **2018**, *18*, 6121–6139.
- (28) Zhang, X.; Yin, Y.; van der A, R.; Eskes, H.; van Geffen, J.; Li, Y.; Kuang, X.; Lapierre, J. L.; Chen, K.; Zhen, Z.; Hu, J.; He, C.; Chen, J.; Shi, R.; Zhang, J.; Ye, X.; Chen, H. Influence of Convection on the Upper-Tropospheric O₃ and NO_x Budget in Southeastern China. *Atmos. Chem. Phys.* **2022**, *22*, 5925–5942.
- (29) Labrador, L. J.; von Kuhlmann, R.; Lawrence, M. G. Strong Sensitivity of the Global Mean OH Concentration and the Tropospheric Oxidizing Efficiency to the Source of NO_x from Lightning. *Geophys. Res. Lett.* **2004**, *31*, L06102.
- (30) Brune, W. H.; McFarland, P. J.; Bruning, E.; Waugh, S.; MacGorman, D.; Miller, D. O.; Jenkins, J. M.; Ren, X.; Mao, J.; Peischl, J. Extreme Oxidant Amounts Produced by Lightning in Storm Clouds. *Science* **2021**, *372*, 711–715.
- (31) Jenkins, J. M.; Brune, W. H.; Miller, D. O. Electrical Discharges Produce Prodigious Amounts of Hydroxyl and Hydroperoxy Radical. *J. Geophys. Res.: Atmos.* **2021**, *126*, e2021JD034557.
- (32) Mao, J.; Zhao, T.; Keller, C. A.; Wang, X.; McFarland, P. J.; Jenkins, J. M.; Brune, W. H. Global Impact of Lightning-Produced Oxidants. *Geophys. Res. Lett.* **2021**, *48*, e2021GL095740.
- (33) Fiore, A. M.; Horowitz, L. W.; Dlugokencky, E. J.; West, J. J. Impact of Meteorology and Emissions on Methane Trends, 1990–2004. *Geophys. Res. Lett.* **2006**, *33*, L12809.
- (34) Murray, L. T. Lightning NO_x and Impacts on Air Quality. *Curr. Pollution. Rep.* **2016**, *2*, 115–133.
- (35) He, J.; Loboda, T. V.; Chen, D.; French, N. H. F. Cloud-to-Ground Lightning and Near-Surface Fire Weather Control Wildfire

- Occurrence in Arctic Tundra. *Geophys. Res. Lett.* **2022**, *49*, e2021GL096814.
- (36) Huntrieser, H.; Schumann, U.; Schlager, H.; Höller, H.; Giez, A.; Betz, H.-D.; Brunner, D.; Forster, C.; Pinto, O.; Calheiros, R. Lightning Activity in Brazilian Thunderstorms during TROCCINOX: Implications for NO_x Production. *Atmos. Chem. Phys.* **2008**, *8*, 921–953.
- (37) Beirle, S.; Huntrieser, H.; Wagner, T. Direct Satellite Observation of Lightning-Produced NO_x. *Atmos. Chem. Phys.* **2010**, *10*, 10965–10986.
- (38) Bucsela, E. J.; Pickering, K. E.; Huntemann, T. L.; Cohen, R. C.; Perring, A.; Gleason, J. F.; Blakeslee, R. J.; Albrecht, R. I.; Holzworth, R.; Cipriani, J. P.; Vargas-Navarro, D.; Mora-Segura, I.; Pacheco-Hernández, A.; Laporte-Molina, S. Lightning-Generated NO_x Seen by the Ozone Monitoring Instrument during NASA's Tropical Composition, Cloud and Climate Coupling Experiment (TC4). *J. Geophys. Res.: Atmos.* **2010**, *115*, 793.
- (39) Allen, D. J.; Pickering, K. E.; Bucsela, E.; Krotkov, N.; Holzworth, R. Lightning NO_x Production in the Tropics as Determined Using OMI NO₂ Retrievals and WWLLN Stroke Data. *J. Geophys. Res. Atmos.* **2019**, *124*, 13498–13518.
- (40) Bucsela, E. J.; Pickering, K. E.; Allen, D.; Holzworth, R.; Krotkov, N. Midlatitude Lightning NO_x Production Efficiency Inferred from OMI and WWLLN Data. *J. Geophys. Res. Atmos.* **2019**, *124*, 13475–13497.
- (41) Said, R. K.; Inan, U. S.; Cummins, K. L. Long-Range Lightning Geolocation Using a VLF Radio Atmospheric Waveform Bank. *J. Geophys. Res.: Atmos.* **2010**, *115*, D23108.
- (42) Said, R. K.; Cohen, M. B.; Inan, U. S. Highly Intense Lightning over the Oceans: Estimated Peak Currents from Global GLD360 Observations. *J. Geophys. Res. Atmos.* **2013**, *118*, 6905–6915.
- (43) Said, R. Towards a Global Lightning Locating System. *Weather* **2017**, *72*, 36–40.
- (44) Vagasky, C. A. Trends in Lightning in the Arctic Region Using GLD360. *Collective Madison Meeting*; 2022.
- (45) Schulz, W.; Cummins, K.; Diendorfer, G.; Dorninger, M. Cloud-to-Ground Lightning in Austria: A 10-Year Study Using Data from a Lightning Location System. *J. Geophys. Res.: Atmos.* **2005**, *110*, D09101.
- (46) Yair, Y.; Shalev, S.; Erlich, Z.; Agrachov, A.; Katz, E.; Saaroni, H.; Price, C.; Ziv, B. Lightning Flash Multiplicity in Eastern Mediterranean Thunderstorms. *Nat. Hazards Earth Syst. Sci.* **2014**, *14*, 165–173.
- (47) Bürgesser, R. E. Assessment of the World Wide Lightning Location Network (WWLLN) Detection Efficiency by Comparison to the Lightning Imaging Sensor (LIS). *Q. J. R. Meteorol. Soc.* **2017**, *143*, 2809–2817.
- (48) Kolmašová, I.; Santolík, O.; Rosická, K. Lightning Activity in Northern Europe during a Stormy Winter: Disruptions of Weather Patterns Originating in Global Climate Phenomena. *Atmos. Chem. Phys.* **2022**, *22*, 3379–3389.
- (49) Yusop, N.; Ahmad, M. R.; Abdullah, M.; Zainudin, S. K.; Nor, W. N. A. W. M.; Alhasa, K. M.; Esa, M. R. M.; Sabri, M. H. M.; Suparta, W.; Gulisano, A. M.; Cooray, V. Cloud-to-Ground Lightning Observations over the Western Antarctic Region. *Polar Sci.* **2019**, *20*, 84–91.
- (50) Mackerras, D.; Darveniza, M. Latitudinal Variation of Lightning Occurrence Characteristics. *J. Geophys. Res. Atmos.* **1994**, *99*, 10813–10821.
- (51) Prentice, S. A.; Mackerras, D. The Ratio of Cloud to Cloud-Ground Lightning Flashes in Thunderstorms. *J. Appl. Meteorol. Climatol.* **1977**, *16*, 545–550.
- (52) Bandholnparat, K.; Sato, M.; Adachi, T.; Ushio, T.; Takahashi, Y. Estimation of the IC to CG Ratio Using JEM-GLIMS and Ground-Based Lightning Network Data. *J. Geophys. Res.: Atmos.* **2020**, *125*, e2019JD032195.
- (53) Christian, H. J.; Blakeslee, R. J.; Boccippio, D. J.; Boeck, W. L.; Buechler, D. E.; Driscoll, K. T.; Goodman, S. J.; Hall, J. M.; Koshak, W. J.; Mach, D. M.; Stewart, M. F. Global Frequency and Distribution of Lightning as Observed from Space by the Optical Transient Detector. *J. Geophys. Res.: Atmos.* **2003**, *108*, ACL 4-1–ACL 4-15.
- (54) Veefkind, J. P.; Aben, I.; McMullan, K.; Förster, H.; de Vries, J.; Otter, G.; Claas, J.; Eskes, H. J.; de Haan, J. F.; Kleipool, Q.; van Weele, M.; Hasekamp, O.; Hoogeveen, R.; Landgraf, J.; Snel, R.; Tol, P.; Ingmann, P.; Voors, R.; Kruizinga, B.; Vink, R.; Visser, H.; Levelt, P. F. TROPOMI on the ESA Sentinel-5 Precursor: A GMES Mission for Global Observations of the Atmospheric Composition for Climate, Air Quality and Ozone Layer Applications. *Remote Sensing of Environment* **2012**, *120*, 70–83.
- (55) van Geffen, J.; Eskes, H.; Compernolle, S.; Pinardi, G.; Verhoelst, T.; Lambert, J.-C.; Sneep, M.; ter Linden, M.; Ludewig, A.; Boersma, K. F.; Veefkind, J. P. Sentinel-5P TROPOMI NO₂ Retrieval: Impact of Version v2.2 Improvements and Comparisons with OMI and Ground-Based Data. *Atmos. Meas. Technol.* **2022**, *15*, 2037–2060.
- (56) Allen, D. J.; Pickering, K. E.; Bucsela, E.; Van Geffen, J.; Lapierre, J.; Koshak, W.; Eskes, H. Observations of Lightning NO_x Production from TROPOMI Case Studies over the United States. *J. Geophys. Res.: Atmos.* **2021**, *126*, e2020JD034174.
- (57) Bäcklund, H.; Hedblom, A.; Neijman, N. A Density-Based Spatial Clustering of Application with Noise. *Data Mining TNM033* **2011**, 11–30.
- (58) Schubert, E.; Sander, J.; Ester, M.; Kriegel, H. P.; Xu, X. DBSCAN Revisited, Revisited: Why and How You Should (Still) Use DBSCAN. *ACM Trans. Database Syst.* **2017**, *42*, 1–21.
- (59) Hersbach, H.; Bell, B.; Berrisford, P.; Hirahara, S.; Horányi, A.; Muñoz-Sabater, J.; Nicolas, J.; Peubey, C.; Radu, R.; Schepers, D.; Simmons, A.; Soci, C.; Abdalla, S.; Abellán, X.; Balsamo, G.; Bechtold, P.; Biavati, G.; Bidlot, J.; Bonavita, M.; Chiara, G.; Dahlgren, P.; Dee, D.; Diamantakis, M.; Dragani, R.; Flemming, J.; Forbes, R.; Fuentes, M.; Geer, A.; Haimberger, L.; Healy, S.; Hogan, R. J.; Hólm, E.; Janisková, M.; Keeley, S.; Laloyaux, P.; Lopez, P.; Lupu, C.; Radnoti, G.; Rosnay, P.; Rozum, I.; Vamborg, F.; Villaume, S.; Thépaut, J.-N. The ERA5 Global Reanalysis. *Q. J. R. Meteorol. Soc.* **2020**, *146*, 1999–2049.
- (60) Soille, P. J.; Ansoult, M. M. Automated Basin Delineation from Digital Elevation Models Using Mathematical Morphology. *Signal Process.* **1990**, *20*, 171–182.
- (61) Heikenfeld, M.; Marinescu, P. J.; Christensen, M.; Watson-Parris, D.; Senf, F.; van den Heever, S. C.; Stier, P. Tobac 1.2: Towards a Flexible Framework for Tracking and Analysis of Clouds in Diverse Datasets. *Geosci. Model Dev.* **2019**, *12*, 4551–4570.
- (62) Lorente, A.; Folkert Boersma, K.; Yu, H.; Dörner, S.; Hilboll, A.; Richter, A.; Liu, M.; Lamsal, L. N.; Barkley, M.; De Smedt, I.; van Roozendael, M.; Wang, Y.; Wagner, T.; Beirle, S.; Lin, J.-T.; Krotkov, N.; Stammes, P.; Wang, P.; Eskes, H. J.; Krol, M. Structural Uncertainty in Air Mass Factor Calculation for NO₂ and HCHO Satellite Retrievals. *Atmos. Meas. Tech.* **2017**, *10*, 759–782.
- (63) Ott, L. E.; Pickering, K. E.; Stenchikov, G. L.; Allen, D. J.; DeCaria, A. J.; Ridley, B.; Lin, R.-F.; Lang, S.; Tao, W.-K. Production of Lightning NO_x and Its Vertical Distribution Calculated from Three-Dimensional Cloud-Scale Chemical Transport Model Simulations. *J. Geophys. Res.: Atmos.* **2010**, *115*, 4711.
- (64) Luo, C.; Wang, Y.; Koshak, W. J. Development of a Self-Consistent Lightning NO_x Simulation in Large-Scale 3-D Models. *J. Geophys. Res. Atmos.* **2017**, *122*, 3141–3154.
- (65) Vasilkov, A.; Joiner, J.; Spurr, R.; Bhartia, P. K.; Levelt, P.; Stephens, G. Evaluation of the OMI Cloud Pressures Derived from Rotational Raman Scattering by Comparisons with Other Satellite Data and Radiative Transfer Simulations. *J. Geophys. Res.: Atmos.* **2008**, *113*, D15S19.
- (66) Joiner, J.; Vasilkov, A. P.; Gupta, P.; Bhartia, P. K.; Veefkind, P.; Sneep, M.; de Haan, J.; Polonsky, I.; Spurr, R. Fast Simulators for Satellite Cloud Optical Centroid Pressure Retrievals; Evaluation of OMI Cloud Retrievals. *Atmos. Meas. Tech.* **2012**, *5*, 529–545.
- (67) Beirle, S.; Salzmann, M.; Lawrence, M. G.; Wagner, T. Sensitivity of Satellite Observations for Freshly Produced Lightning NO_x. *Atmos. Chem. Phys.* **2009**, *9*, 1077–1094.

- (68) Laughner, J. L.; Cohen, R. C. Quantification of the Effect of Modeled Lightning NO₂ on UV-Visible Air Mass Factors. *Atmos. Meas. Technol.* **2017**, *10*, 4403–4419.
- (69) Frey, W.; Schofield, R.; Hoor, P.; Kunkel, D.; Ravagnani, F.; Ulanovsky, A.; Viciani, S.; D'Amato, F.; Lane, T. P. The Impact of Overshooting Deep Convection on Local Transport and Mixing in the Tropical Upper Troposphere/Lower Stratosphere (UTLS). *Atmos. Chem. Phys.* **2015**, *15*, 6467–6486.
- (70) Pickering, K. E.; Bucsela, E.; Allen, D.; Ring, A.; Holzworth, R.; Krotkov, N. Estimates of Lightning NO_x Production Based on OMI NO₂ Observations over the Gulf of Mexico. *J. Geophys. Res. Atmos.* **2016**, *121*, 8668–8691.
- (71) Lapierre, J. L.; Laughner, J. L.; Geddes, J. A.; Koshak, W.; Cohen, R. C.; Pusede, S. E. Observing U.S. Regional Variability in Lightning NO₂ Production Rates. *J. Geophys. Res.: Atmos.* **2020**, *125*, e2019JD031362.
- (72) Zhang, X.; Yin, Y.; van der A, R.; Lapierre, J. L.; Chen, Q.; Kuang, X.; Yan, S.; Chen, J.; He, C.; Shi, R. Estimates of Lightning NO_x Production Based on High-Resolution OMI NO₂ Retrievals over the Continental US. *Atmos. Meas. Tech.* **2020**, *13*, 1709–1734.
- (73) Pérez-Invernón, F. J.; Huntrieser, H.; Erbertseder, T.; Loyola, D.; Valks, P.; Liu, S.; Allen, D. J.; Pickering, K. E.; Bucsela, E. J.; Jöckel, P.; van Geffen, J.; Eskes, H.; Soler, S.; Gordillo-Vázquez, F. J.; Lapierre, J. Quantification of Lightning-Produced NO_x over the Pyrenees and the Ebro Valley by Using Different TROPOMI-NO₂ and Cloud Research Products. *Atmos. Meas. Technol.* **2022**, *15*, 3329–3351.
- (74) McCarty, J. L.; Aalto, J.; Paunu, V.-V.; Arnold, S. R.; Eckhardt, S.; Klimont, Z.; Fain, J. J.; Evangelou, N.; Venäläinen, A.; Tchebakova, N. M.; Parfenova, E. I.; Kupiainen, K.; Soja, A. J.; Huang, L.; Wilson, S. Reviews and Syntheses: Arctic Fire Regimes and Emissions in the 21st Century. *Biogeosciences* **2021**, *18*, 5053–5083.
- (75) Schmale, J.; Arnold, S. R.; Law, K. S.; Thorp, T.; Anenberg, S.; Simpson, W. R.; Mao, J.; Pratt, K. A. Local Arctic Air Pollution: A Neglected but Serious Problem. *Earth's Future* **2018**, *6*, 1385–1412.
- (76) Jacob, D. J.; Crawford, J. H.; Maring, H.; Clarke, A. D.; Dibb, J. E.; Emmons, L. K.; Ferrare, R. A.; Hostetler, C. A.; Russell, P. B.; Singh, H. B.; Thompson, A. M.; Shaw, G. E.; McCauley, E.; Pederson, J. R.; Fisher, J. A. The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) Mission: Design, Execution, and First Results. *Atmos. Chem. Phys.* **2010**, *10*, 5191–5212.
- (77) Network, V. L. D.Total Lightning Statistics 2021: Vaisala Annual Lightning Report. 2021.
- (78) van der A, R. J.; de Laat, A. T. J.; Ding, J.; Eskes, H. J. Connecting the Dots: NO_x Emissions along a West Siberian Natural Gas Pipeline. *npj Clim. Atmos. Sci.* **2020**, *3*, 16.
- (79) Hachmeister, J.; Schneising, O.; Buchwitz, M.; Lorente, A.; Borsdorff, T.; Burrows, J. P.; Notholt, J.; Buschmann, M. On the Influence of Underlying Elevation Data on Sentinel-5 Precursor TROPOMI Satellite Methane Retrievals over Greenland. *Atmos. Meas. Technol.* **2022**, *15*, 4063–4074.
- (80) Goldberg, D. L.; Anenberg, S. C.; Kerr, G. H.; Mohegh, A.; Lu, Z.; Streets, D. G. TROPOMI NO₂ in the United States: A Detailed Look at the Annual Averages, Weekly Cycles, Effects of Temperature, and Correlation With Surface NO₂ Concentrations. *Earth's Future* **2021**, *9*, e2020EF001665.
- (81) Marais, E. A.; Jacob, D. J.; Choi, S.; Joiner, J.; Belmonte-Rivas, M.; Cohen, R. C.; Beirle, S.; Murray, L. T.; Schiferl, L. D.; Shah, V.; Jaeglé, L. Nitrogen Oxides in the Global Upper Troposphere: Interpreting Cloud-Sliced NO₂ Observations from the OMI Satellite Instrument. *Atmos. Chem. Phys.* **2018**, *18*, 17017–17027.
- (82) Beirle, S.; Koshak, W.; Blakeslee, R.; Wagner, T. Global Patterns of Lightning Properties Derived by OTD and LIS. *Nat. Hazards Earth Syst. Sci.* **2014**, *14*, 2715–2726.
- (83) Hutchins, M. L.; Holzworth, R. H.; Virt, K. S.; Wallace, J. M.; Heckman, S. Radiated VLF Energy Differences of Land and Oceanic Lightning. *Geophys. Res. Lett.* **2013**, *40*, 2390–2394.
- (84) Bruning, E. C.; MacGorman, D. R. Theory and Observations of Controls on Lightning Flash Size Spectra. *J. Atmos. Sci.* **2013**, *70*, 4012–4029.
- (85) Bruning, E. C.; Thomas, R. J. Lightning Channel Length and Flash Energy Determined from Moments of the Flash Area Distribution. *J. Geophys. Res. Atmos.* **2015**, *120*, 8925–8940.
- (86) Mecikalski, R. M.; Bain, A. L.; Carey, L. D. Radar and Lightning Observations of Deep Moist Convection across Northern Alabama during DC3:21 May 2012. *Mon. Wea. Rev.* **2015**, *143*, 2774–2794.
- (87) Silvern, R. F.; Jacob, D. J.; Travis, K. R.; Sherwen, T.; Evans, M. J.; Cohen, R. C.; Laughner, J. L.; Hall, S. R.; Ullmann, K.; Crounse, J. D.; Wennberg, P. O.; Peischl, J.; Pollack, I. B. Observed NO/NO₂ Ratios in the Upper Troposphere Imply Errors in NO-NO₂-O₃ Cycling Kinetics or an Unaccounted NO_x Reservoir. *Geophys. Res. Lett.* **2018**, *45*, 4466–4474.
- (88) Belmonte Rivas, M.; Veefkind, P.; Eskes, H.; Levelt, P. OMI Tropospheric NO₂ Profiles from Cloud Slicing: Constraints on Surface Emissions, Convective Transport and Lightning NO_x. *Atmos. Chem. Phys.* **2015**, *15*, 13519–13553.
- (89) Marais, E. A.; Roberts, J. F.; Ryan, R. G.; Eskes, H.; Boersma, K. F.; Choi, S.; Joiner, J.; Abuahassan, N.; Redondas, A.; Grutter, M.; Cede, A.; Gomez, L.; Navarro-Comas, M. New Observations of NO₂ in the Upper Troposphere from TROPOMI. *Atmos. Meas. Technol.* **2021**, *14*, 2389–2408.
- (90) Barth, M. C.; Rutledge, S. A.; Brune, W. H.; Cantrell, C. A. Introduction to the Deep Convective Clouds and Chemistry (DC3) 2012 Studies. *J. Geophys. Res. Atmos.* **2019**, *124*, 8095–8103.
- (91) Law, K. S.; Stohl, A. Arctic Air Pollution: Origins and Impacts. *Science* **2007**, *315*, 1537–1540.
- (92) Cecil, D. J.; Buechler, D. E.; Blakeslee, R. J. Gridded Lightning Climatology from TRMM-LIS and OTD: Dataset Description. *Atmos. Res.* **2014**, *135–136*, 404–414.
- (93) Blakeslee, R. J.; Lang, T. J.; Koshak, W. J.; Buechler, D.; Gatlin, P.; Mach, D. M.; Stano, G. T.; Virts, K. S.; Walker, T. D.; Cecil, D. J.; Ellett, W.; Goodman, S. J.; Harrison, S.; Hawkins, D. L.; Heumesser, M.; Lin, H.; Maskey, M.; Schultz, C. J.; Stewart, M.; Bateman, M.; Chanrior, O.; Christian, H. Three Years of the Lightning Imaging Sensor Onboard the International Space Station: Expanded Global Coverage and Enhanced Applications. *J. Geophys. Res.: Atmos.* **2020**, *125*, e2020JD032918.
- (94) Goodman, S. J.; Blakeslee, R. J.; Koshak, W. J.; Mach, D.; Bailey, J.; Buechler, D.; Carey, L.; Schultz, C.; Bateman, M.; McCaul, E.; Stano, G. The GOES-R Geostationary Lightning Mapper (GLM). *Atmos. Res.* **2013**, *125–126*, 34–49.
- (95) Yang, J.; Zhang, Z.; Wei, C.; Lu, F.; Guo, Q. Introducing the New Generation of Chinese Geostationary Weather Satellites, Fengyun-4. *Bull. Am. Meteor. Soc.* **2017**, *98*, 1637–1658.
- (96) Asmus, V. V.; Milekhin, O. E.; Kramareva, L. S.; Khailov, M. N.; Shirshakov, A. E.; Shumakov, I. A. Arktika-M: The World's First Highly Elliptical Orbit Hydrometeorological Space System. *Russ. Meteorol. Hydrol.* **2021**, *46*, 805–816.
- (97) Zhang, X.Zxdawn/SSP-LNO2, ver. 1.0; Zenodo, 2023.
- (98) Zhang, X.Zxdawn/SSP-LNO2-Notebook, ver. 1.0; Zenodo, 2023.
- (99) Zhang, X. Data set for Spaceborne Observations of Lightning NO₂ in the Arctic. 2023.