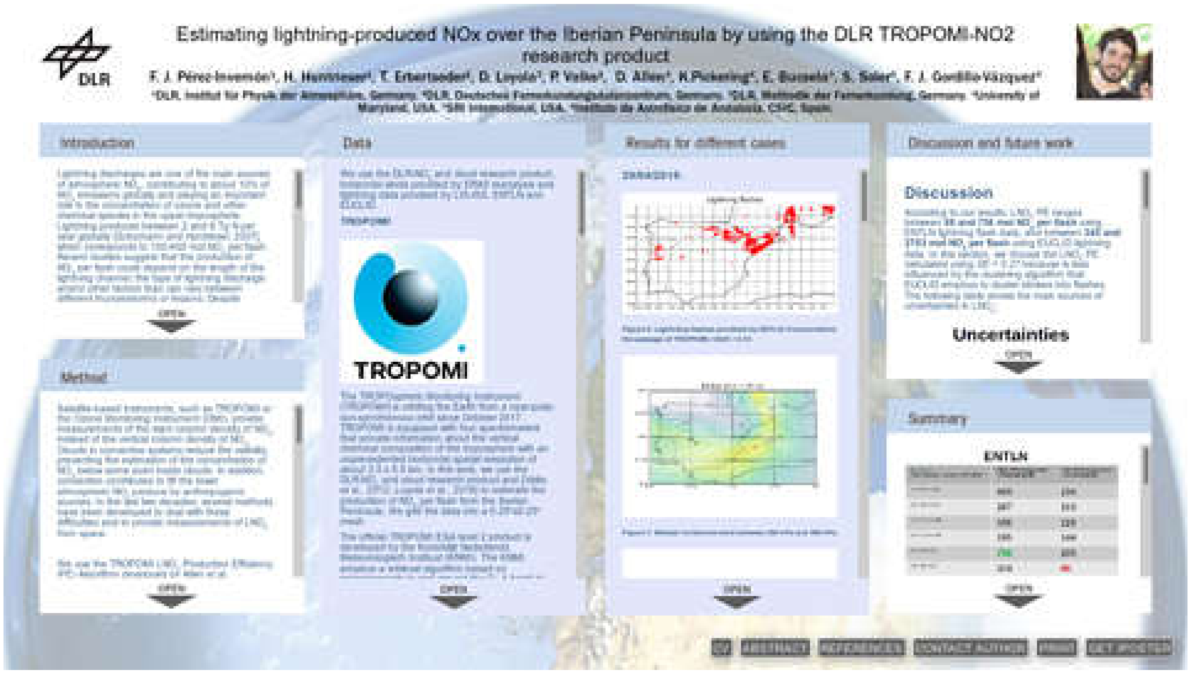
Estimating lightning-produced NOx over the Iberian Peninsula by using the DLR TROPOMI-NO2 research product



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INTRODUCTION

Lightning discharges are one of the main sources of atmospheric NO_x, contributing to about 10% of NO_x emissions globally and playing an important role in the concentration of ozone and other chemical species in the upper troposphere. Lightning produces between 2 and 8 Tg N per year globally [Schumann and Huntrieser, 2007], which corresponds to 100-400 mol NO_x per flash. Recent studies suggest that the production of NO_x per flash could depend on the length of the lightning channel, the type of lightning discharge and/or other factors than can vary between different thunderstorms or regions. Despite significant advances achieved by aircraft campaigns and by the improvement of satellites during the last two decades, reducing the uncertainty in the production of NO_x by lightning and understanding the factors that influence the production in different thunderstorms is still a challenge.

The TROPOspheric Monitoring Instrument (*TROPOMI*) is orbiting the Earth from a near-polar, sun-synchronous orbit since October 2017. TROPOMI is equipped with four spectrometers that provide information about the vertical chemical composition of the troposphere with an unprecedented horizontal spatial resolution of about 3.5 x 5.5 km. In this work, we use the DLR-NO_2 research product and the DLR cloud operational product to estimate the production of NO_x per flash (LNO_x) from the Iberian Peninsula. We for the first time ever use chemical measurements from TROPOMI combined with lightning radio measurements provided by the EUropean Cooperation for LIghtning Detection (EUCLID) and the Earth Network Total Lightning Network (ENTLN), together with lightning optical measurements provided by the space-based Lightning Imaging Sensor (LIS) to estimate the Detection Effiency (DE) of EUCLID and ENTLN.

One of the main sources of uncertainty in the estimation of LNO_x is the influence of the background concentration of NO_x. The source of tropospheric background can be either anthropogenic emissions that have been convectively lofted to the upper troposphere or LNO_x from upwind storms. We have considered the winds provided by reanalysis data to reduce the influence of the upwind storms in the background.

We focus our analysis in different regions of the Iberian Peninsula, where the background concentration of NO is relatively low. In particular, we focus our analysis on cases of thunderstorms taking place near the Pyrenees, where the background concentration of NO_x is low, active thunderstorms are frequent and the DE of EUCLID and ENTLN is relatively high and homogeneous.

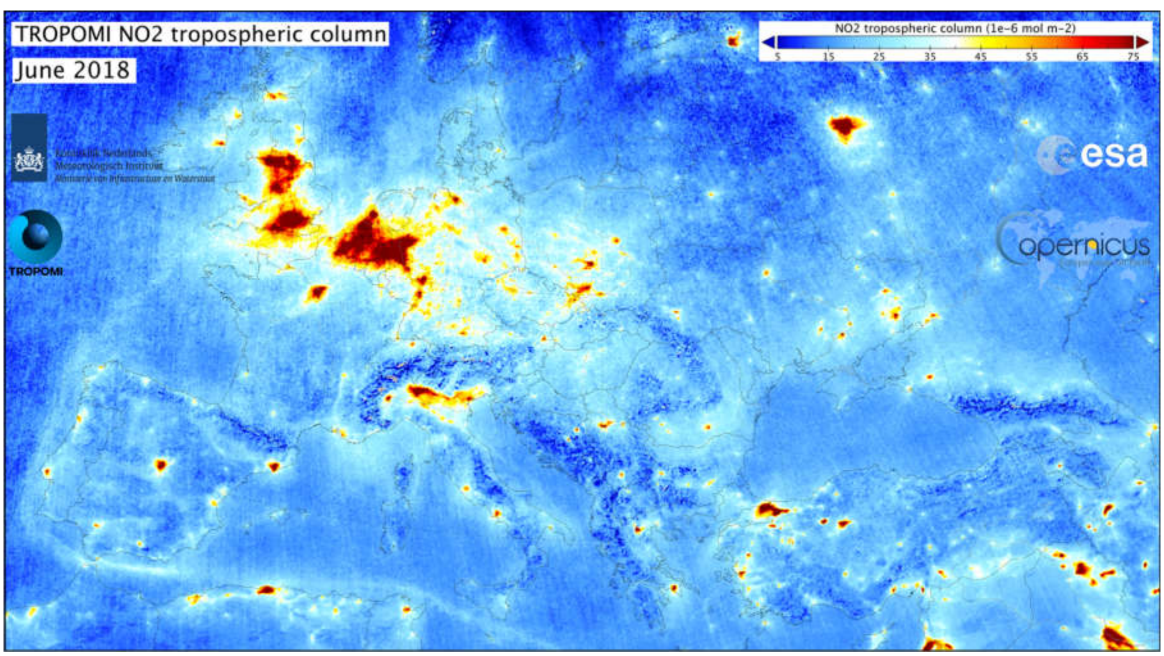

Figure 1: TROPOMI Nitrogen Dioxide (NO_2) tropospheric column density provided by the European Space Agency (ESA).

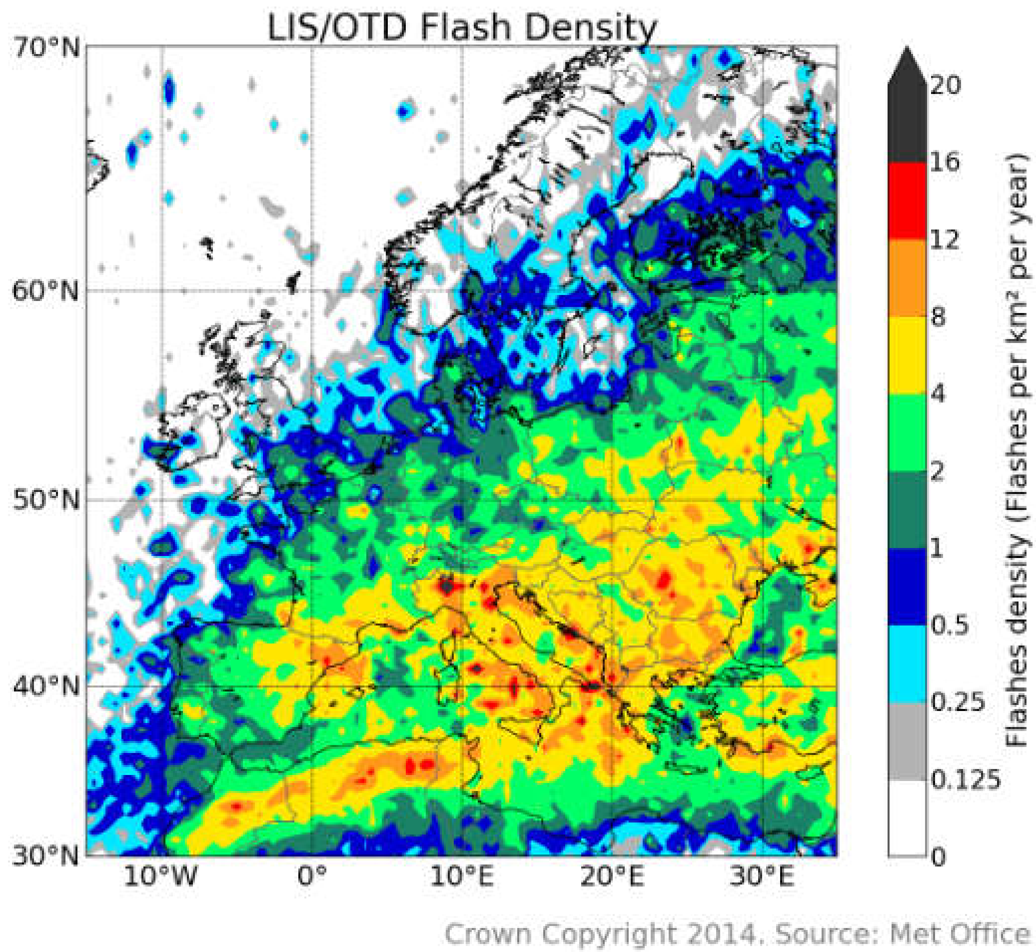

Figure 2:

Estimated European flash density obtained using data from the OTD and LIS satellite-based instruments. Adapted from Anderson and Klugmann (2014).

METHOD

Satellite-based instruments, such as TROPOMI or the Ozone Monitoring Instrument (OMI), provide measurements of the slant column density of NO_2 instead of the vertical column density of NO_x. Clouds in convective systems reduce the visibility, preventing the estimation of the concentration of NO_2 below some point inside clouds. In addition, convection contributes to lift the lower atmospheric NO_x produce by anthropogenic sources. In the last two decades, several methods have been developed to deal with these difficulties and to provide measurements of LNO_x from space.

We use the TROPOMI LNO_x Production Efficiency (PE) Algorithm developed by Allen et al. (AGU-2019), that is based on the OMI LNO_x PE Algorithm employed in previous studies to estimate the fresh NO_x produced by lightning prior to the passage of the instrument [e.g., Pickering et al., (2016), Bucsela et al., (2019) and Allen et al., (2019)]. In this section, we describe the algorithm and discuss the choice of the employed parameters.

The LNO_x Production Efficiency (moles NO_x per flash) is calculated according to

$$PE = \frac{V_{tropLNOx} \times A}{N_A \times \Sigma(F\ times\, \exp(-t/\tau))},$$

where

$\boldsymbol{V_{tropLNOx}}$ is the median vertical column density (VCD) of LNO_x over pixels that satisfy the Deep Convective Constraint (DCC, see below),

$\boldsymbol{A}$ is the area of pixels that satisfy DCC,

$\boldsymbol{N_A}$ is the Avogadro's number,

$\boldsymbol{F}$ is the total number of ENTLN or EUCLID flashes during 1 or 5 hours before TROPOMI overpass time,

$\boldsymbol{t}$ is the age of individual flashes at the time of the TROPOMI overpass and

$\boldsymbol{\tau}$ is the lifetime of NO_2 in near field of convection (between 3 and 12 hours).

The tropospheric NO_x produced by lightning ($V_{tropLNOx}$) is calculated as the difference of the tropospheric VCD of NO_x ($V_{tropNOx}$) and the background ($V_{tropbck}$) according to:

$$V_{tropLNOx} = Median(V_{tropNOx}) - V_{tropbck}.$$

However, TROPOMI does not provide measurements of $V_{tropNOx}$. This quantiy is estimated from the **TROPOMI DLR-NO_2 research product** variables: 1) NO_2 slant column density (S_{NO2}), 2) stratospheric VCD of NO_2 ($V_{stratNO2}$) and 3) stratospheric air mass factor (AMF_{strat}) over DCC pixels according to

$$V_{tropNOx} = \frac{S_{NO2} - avg(V_{stratNO2} \times AMF_{strat})}{AMF_{LNOx}},$$

where AMF_{LNOx} is the AMF converting tropospheric slant column of NO_2 into vertical column of LNO_x. This parameter is usually calculated using atmospheric models and scattering weights. However, in this preliminary study, we will consider that it can vary between 0.3 and 0.7 according to Beirle et al., (2009).

The estimation of the background tropospheric NO_x is one of the main sources of uncertainty in the final value of PE. In this preliminary study, we estimate $V_{tropbck}$ as the $10^{th}(40^{th})\%$ of $V_{tropNOx}$ for non-flashing pixels satisfying DCC [Allen et al., AGU-2019]. We exclude from the background the pixels around the flash positions. We use the median value and direction of the horizontal winds between 200 hPa and 500 hPa to exclude the cells that are likely to have been influenced by LNO_x.

We use the Deep Convective Constraint (DCC) to extract pixels with cloud fraction > 0.95 [Allen et al., AGU-2019] and cloud pressure lower than a given value (593 hPa). We have estimated the typical cloud pressure for lightning in the Iberian Peninsula by collecting the DLR-NO_2 research product Optical Centroid Pressure (OCP) value of each lightning flash included in this study, finding a mean value of 593 hPa (see Fig. 3). Pickering et al., (2016) used a criterion of 650 hPa for the OCP, whereas Allen et al., (2019) and Bucsela et al., (2019) used 500 hPa.

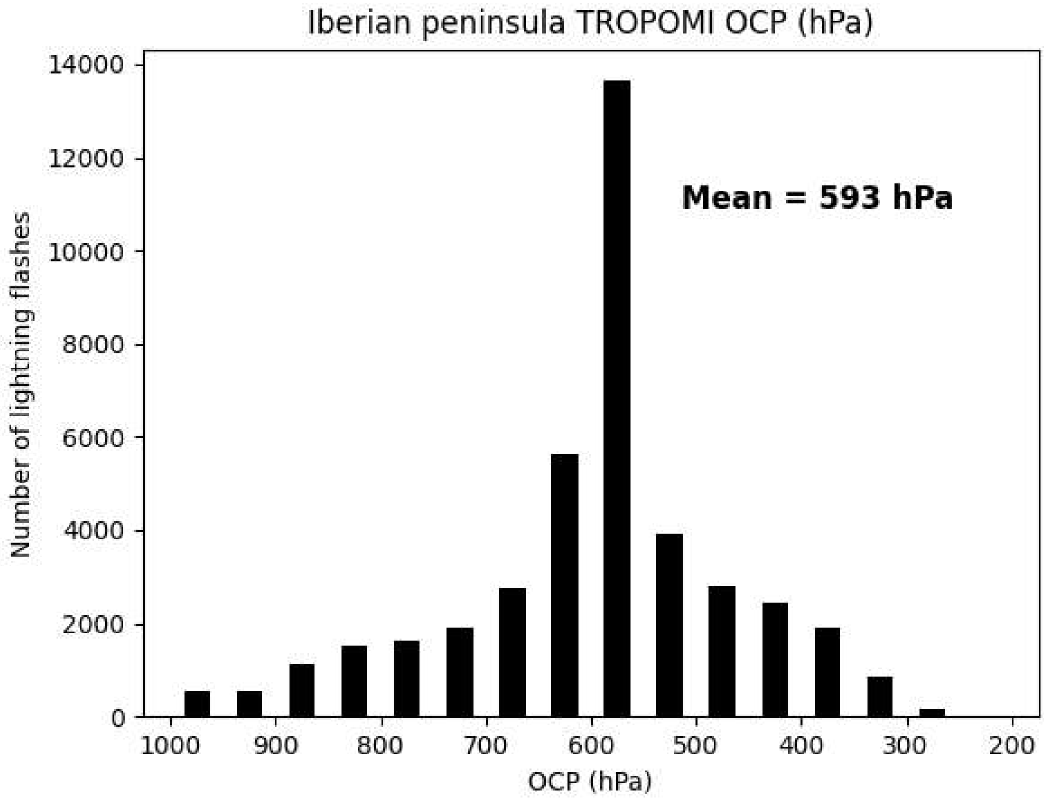

Figure 3: Distribution of the Optical Centroid Pressure (OCP) values for all the lightning flashes included in this study provided by the DLR-NO_2 research product.

DATA

We use the DLR-NO_2 and cloud research product, horizontal winds provided by ERA5 reanalysis and lightning data provided by LIS-ISS, ENTLN and EUCLID.

TROPOMI

The TROPOspheric Monitoring Instrument (*TROPOMI*) is orbiting the Earth from a near-polar, sun-synchronous orbit since October 2017. TROPOMI is equipped with four spectrometers that provide information about the vertical chemical composition of the troposphere with an unprecedented horizontal spatial resolution of about 3.5 x 5.5 km. In this work, we use the DLR-NO_2 and cloud research product and [Valks et al., 2012, Loyola et al., 2018] to estimate the production of NO_x per flash from the Iberian Peninsula. We grid the data into a 0.25ºx0.25º mesh.

The official TROPOMI ESA level 2 product is developed by the Koninklijk Nederlands Meteorologisch Instituut (KNMI). The KNMI employs a retrieval algorithm based on measurements in and around the O_2 A-band at 760 nm called Fast REtrieval Scheme for Clouds from the Oxygen A-band (FRESCO). The FRESCO algorithm is based on the calculation of transmittances and retrieves effective cloud fraction and cloud top pressure, **assuming a fixed cloud albedo of 0.8**. The DLR-NO_2 research product (used in this work) combines the OCRA (Optical Cloud Recognition Algorithm) and the ROCINN (Retrieval of Cloud Information using Neural Networks) [Loyola et al. (2018)]. OCRA retrieves the cloud fraction using TROPOMI measurements in the ultraviolet (UV) and visible (VIS) spectral regions, while ROCINN retrieves the cloud top height (pressure) and optical thickness (**albedo**) using TROPOMI measurements in and around the oxygen A-band in the near infrared (NIR). The calculation of the albedo makes the use of the TROPOMI DLR product suitable for calculating LNO_x over bright clouds.

LIS-ISS

The Lightning Imaging Sensor (LIS) was placed on the International Space Station (ISS) for a two-four year mission starting in March 2017 covering the range of latitude between 54.3ºN and 54.3ºS [Blakeslee et al., 2020]. LIS detects optical emissions from lightning with a frame integration time of 1.79 ms. LIS groups contiguous events into groups, and clusters groups into flashes with a temporal criteria of 330 ms and an spatial criteria of 5.5 km.

The Detection Efficiency (DE) of LIS-ISS over the Iberian Peninsula ranges between 70 and 100% [Poelman and Schultz, 2020], We use the lightning data provided by LIS-ISS to estimate the DE of ground-based Lightning Location Systems (LLS).

ENTLN

EARTH NETWORKS

The ground-based Earth Network Total Lightning Network (ENTLN) is a global network composed of Very Low Frequency (VLF) sensors that provide the position, time of occurrence, polarity and peak current of lightning strokes. We use lightning flash data provided by ENTLN to calculate the production of NO_x per flash. ENTLN has a DE of about 90% for CG strokes, between 44% and 63% for IC strokes over the US [Zhu et al., 2017, Lapierre et al., 2020] and a total global stroke DE of about 57% [Bitzer and Burchfield, (2016)]. We use the LIS-ISS lightning data together with the Bayesian technique proposed by Bitzer and Burchfield (2016) to estimate the total flash DE of ENTLN between 2017 and 2018 over the Iberian Peninsula. In the comparison, we take into that the flash criteria proposed by Liu et al. 2011 to cluster ENTLN strokes into ENTLN flashes is 0.7 s and 10 km.

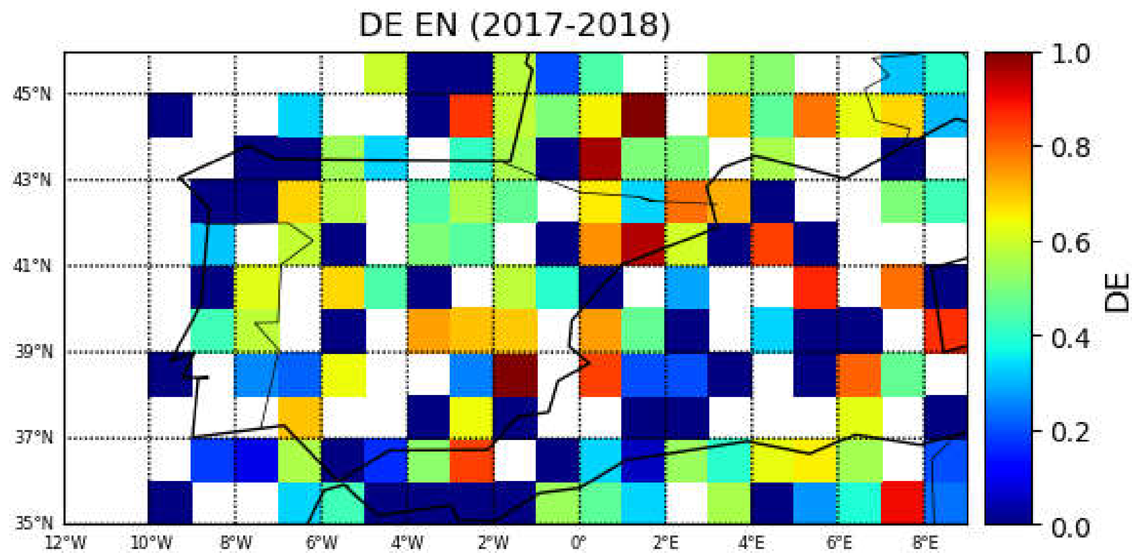

Figure 4: Total flash DE of ENTLN calculated using LIS-ISS data accoring to the Bayesian technique proposed by Bitzer and Burchfield, (2016).

EUCLID

EUCLID
European Cooperation for Lightning Detection

The ground-based European Cooperation for Lightning Detection (EUCLID) is an European network composed of VLF sensors that provide the position, time of occurrence, polarity and peak current of lightning strokes. We use lightning flash data provided by EUCLID between April and May 2018 to calculate the production of NOx per flash.

Poelman and Schulz (2020) investigated the DE of EUCLID between 2017 and 2019 over the Iberian Peninsula. According to their results, EUCLID flash DE over the Pyrenees could range between 0.4 and 0.5. However, the calculated flash DE can be influenced by the clustering algorithms of the lightning detection systems to cluster strokes into flashes. Therefore, we have estimated both the flash and the stroke DE of EUCLID over the Pynerees using a thunderstorm that was simultaneously detected by EUCLID and LIS-ISS on April 27, 2018. The calculation of the DE using strokes with peak current absolute value greater than 10 kA instead of flashes is not influenced by the EUCLID clustering algorithm.

LIS-ISS detected 29 flashes during its passage over the thunderstorm, while EUCLID reported 13 flashes, among which there were 8 strokes with peak current absolute value greater than 10 kA.

Based on these numbers, we use EUCLID DE values of 0.5 (flash DE) and 0.27 (stroke DE) to estimate the LNOx.

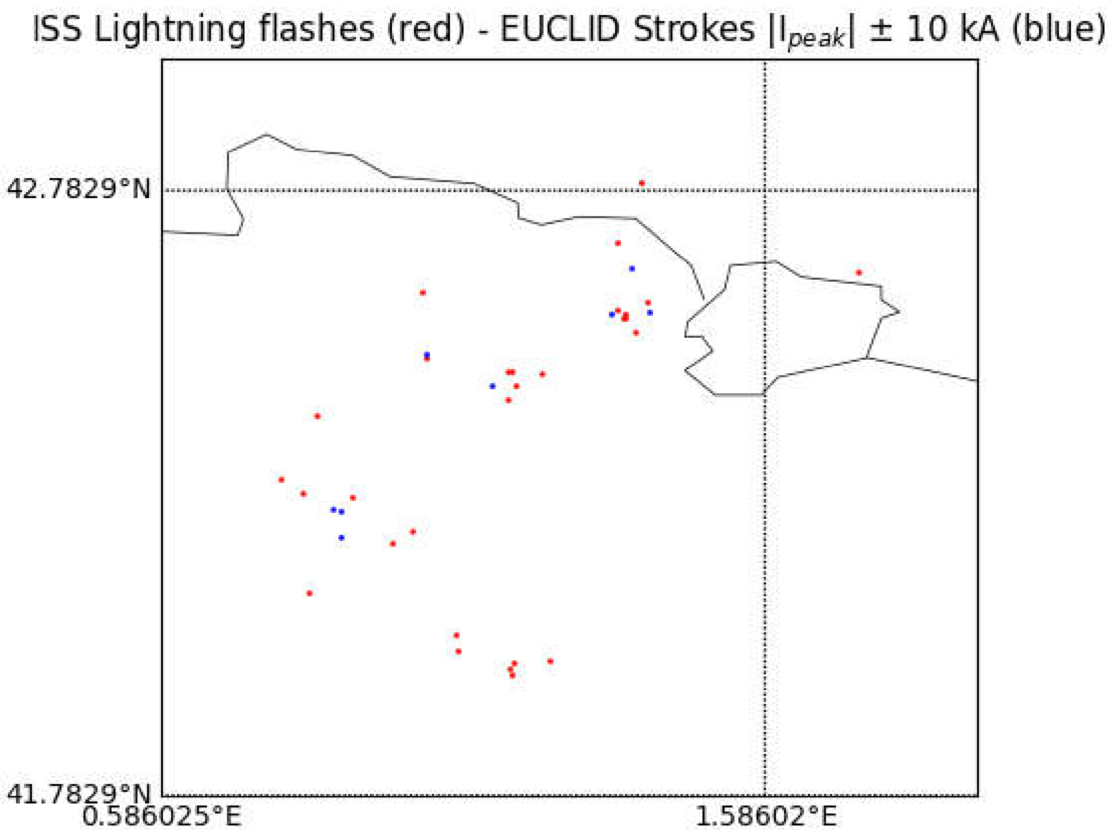

Figure 5: Flashes reported by LIS-ISS (red dots) and strokes with $|I_{peak}|$>10 kA reported by EUCLID.

ERA5

ECMWF

The European Centre for Medium-Range Weather Forecasts (ECMWF) fifth generation reanalysis (ERA5) provides 1-hourly meteorological data using a 4D-var assimilation scheme at 139 pressure levels with an horizontal resolution of 0.25º. We use the horizontal winds between 200 hPa and 500 hPa levels to estimate the spreading of the LNO_x before the passage of TROPOMI.

RESULTS FOR DIFFERENT CASES

29/04/2018:

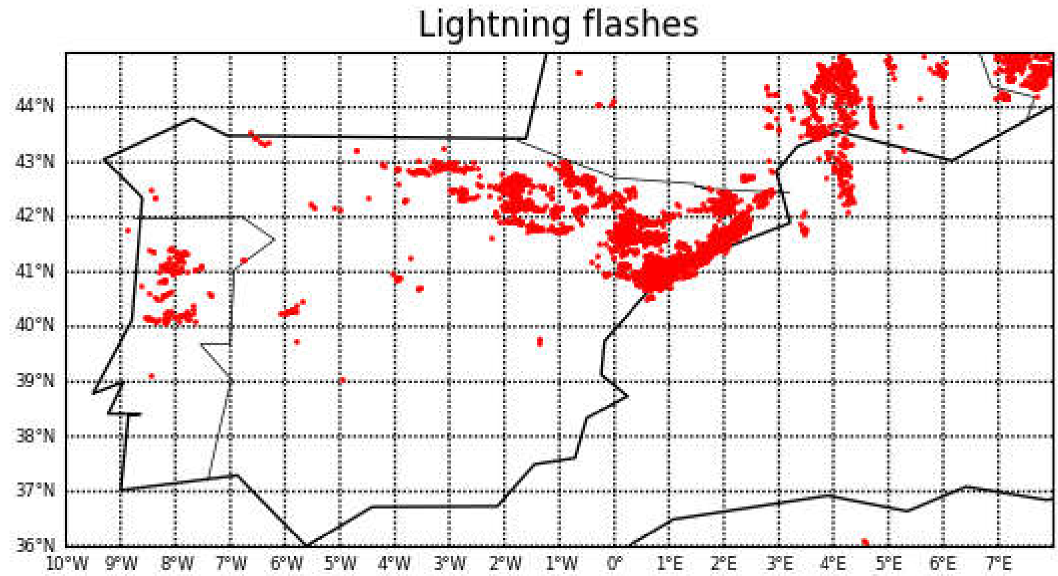

Figure 6: Lightning flashes provided by ENTLN 5 hours before the passage of TROPOMI. **CG/IC = 0.72**

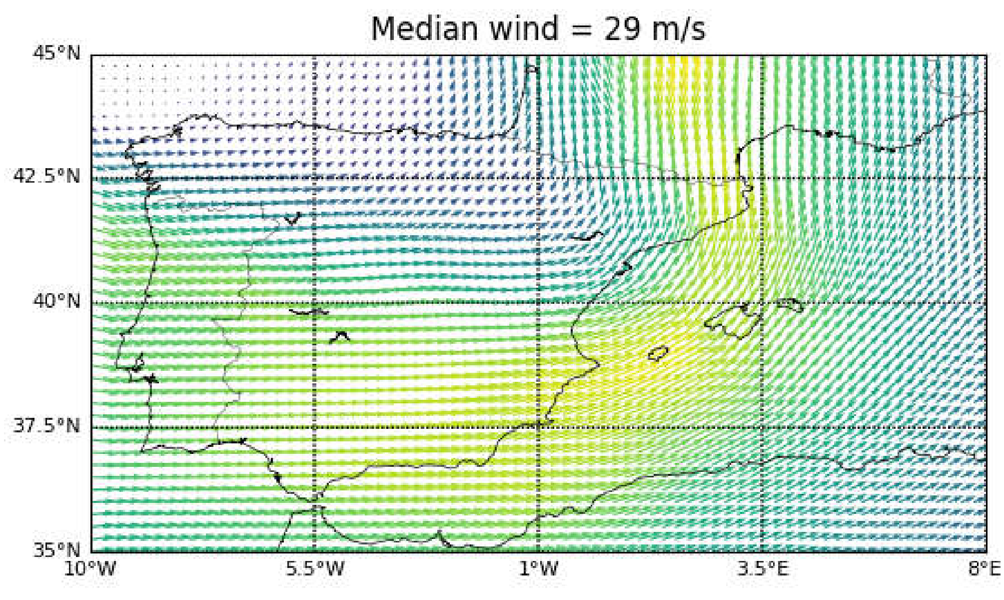

Figure 7: Median horizontal wind between 200 hPa and 500 hPa.

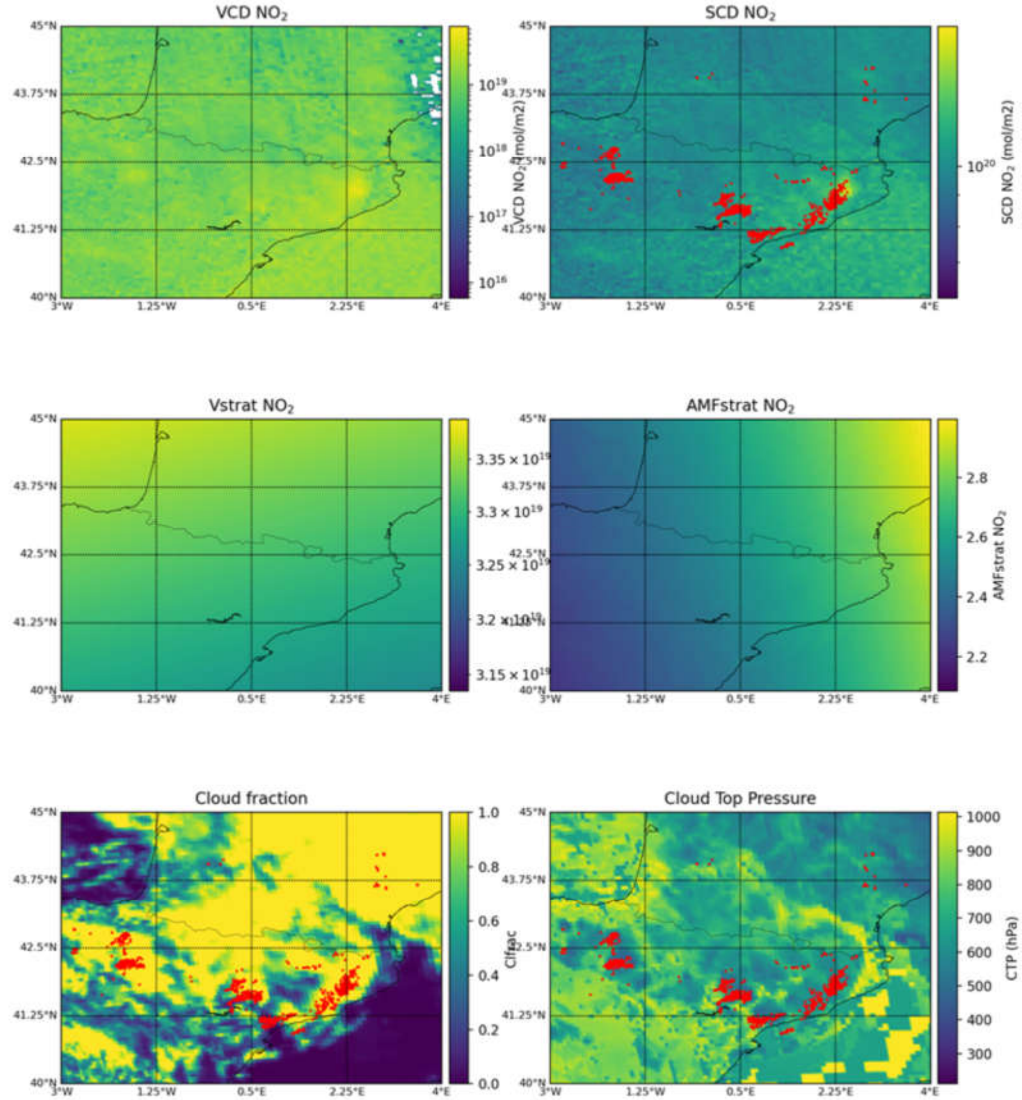

Figure 8: TROPOMI DLR-NO_2 product and flashes 1 hour before the passage of TROPOMI (red dots).

Parameters Flash Window / Lifetime / AMFLNOX	Nflashes	Age	LNOx 10 th background - No corrected by DE	NOx 40 th background - No corrected by DE	LNOx 10 th background - Corrected by DE = 0.5	LNOx 40 th background - Corrected by DE = 0.5	**LNOx 10 th background - Corrected by DE = 0.27**	**LNOx 40 th background - Corrected by DE = 0.27**
1 h / 3 h / 0.46	262	0.53 h	9979	2280	4985	1140	**2694**	**616**
5 h / 3 h / 0.46	849	1.95 h	5112	4079	2556	2039	**1380**	**1101**
1 h / 12 h / 0.46	262	0.53 h	8763	2003	4381	1001	**2366**	**541**
5 h / 12 h / 0.46	849	1.95 h	3402	2715	1701	1357	**918**	**733**
1 h / 3 h / 0.3	262	0.53 h	15301	3497	7650	1748	**4131**	**9446**
1 h / 3 h / 0.7	262	0.53 h	6558	1499	3279	749	**1771**	**405**

Table 1: LNO_x PE (mol NO_x per flash) using different parameters and EUCLID lightning data.

Parameters Flash Window / Lifetime / AMFLNOX	Nflashes	Age	LNOx 10 th background - No corrected by DE	NOx 40 th background - No corrected by DE	**LNOx 10 th backgroud - Corrected by DE**	**LNOx 40 th backgroud - Corrected by DE**
1 h / 3 h / 0.46	1331	0.56 h	1989	522	**1168**	**306**
5 h / 3 h / 0.46	4129	1.94 h	1044	815	**635**	**496**
1 h / 12 h / 0.46	1331	0.56 h	1735	455	**1026**	**269**
5 h / 12 h / 0.46	4129	1.94 h	695	542	**419**	**327**
1 h / 3 h / 0.3	1331	0.56 h	3051	800	**1792**	**470**
1 h / 3 h / 0.7	1331	0.56 h	1308	343	**768**	**201**

Table 2: LNO_x PE (mol NO_x per flash) using different parameters and ENTLN lightning data.

12/05/2018:

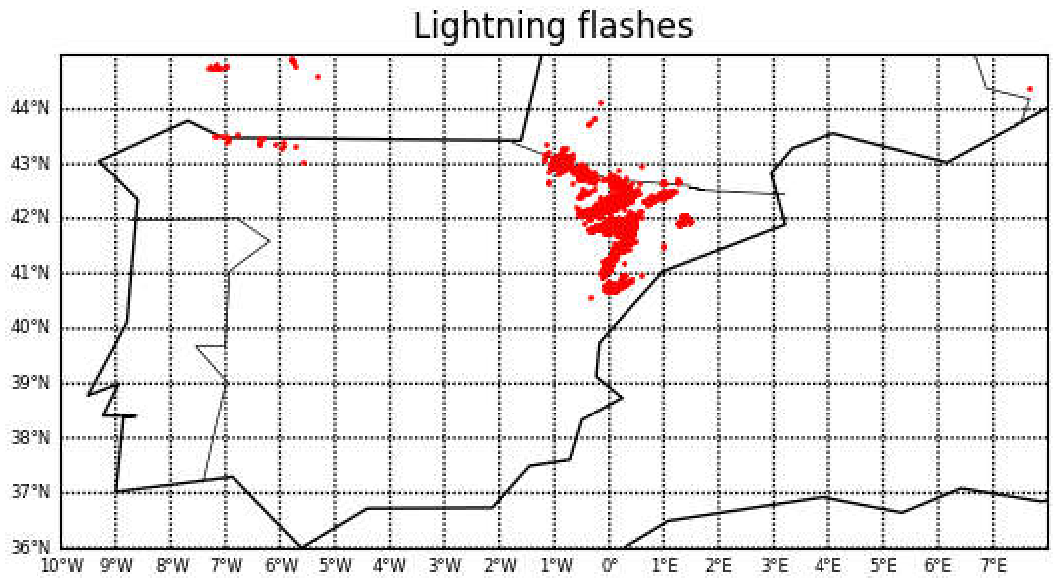

Figure 9: Lightning flashes provided by ENTLN 5 hours before the passage of TROPOMI.

CG/IC = 0.10

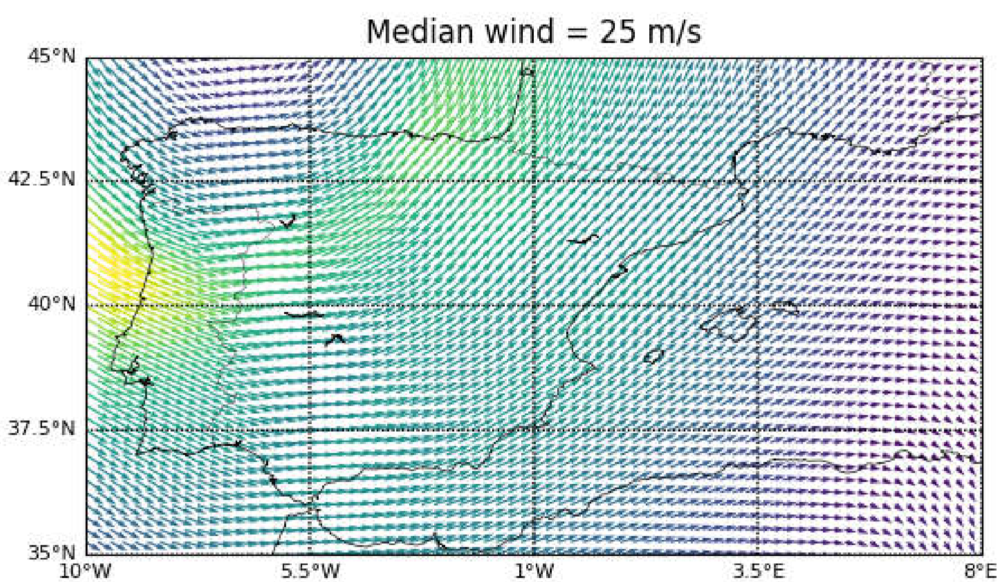

Figure 10: Median horizontal wind between 200 hPa and 500 hPa.

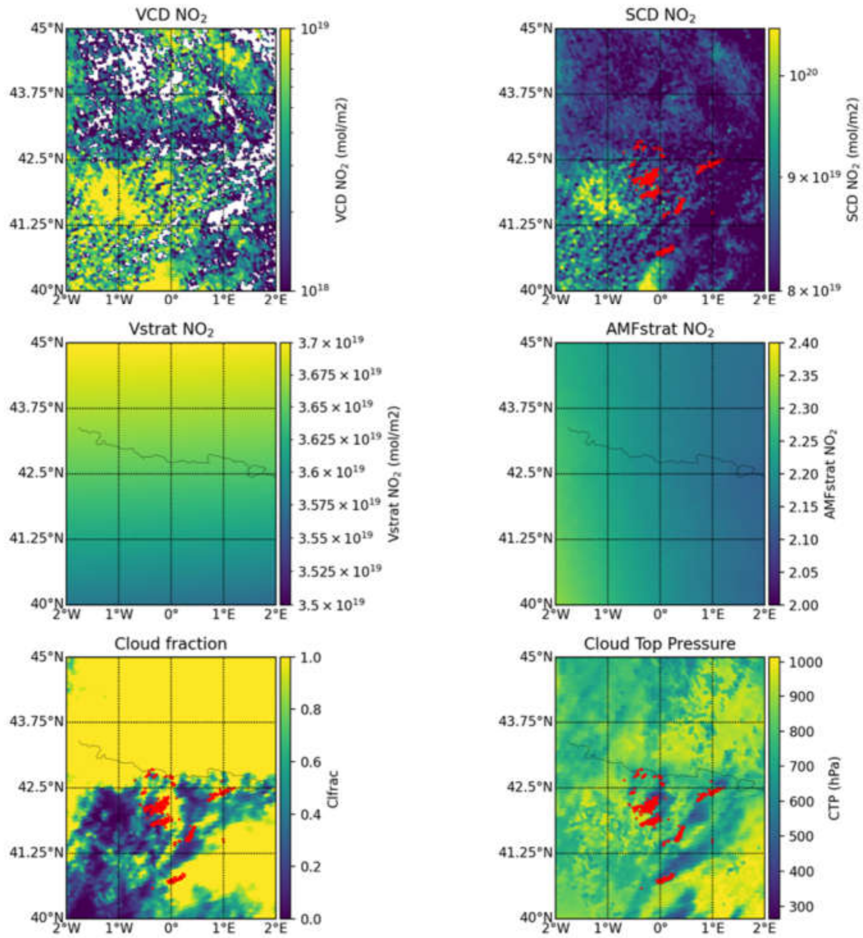

Figure 11: TROPOMI DLR-NO$_2$ product and flashes 1 hour before the passage of TROPOMI (red dots).

Parameters Flash Window / Lifetime / AMFLNOX	Nflashes	Age	LNOx 10 th background - No corrected by DE	NOx 40 th background - No corrected by DE	LNOx 10 th background - Corrected by DE = 0.5	LNOx 40 th background - Corrected by DE = 0.5	**LNOx 10 th background - Corrected by DE = 0.27**	**LNOx 40 th background - Corrected by DE = 0.27**
1 h / 3 h / 0.46	73	0.39 h	3079	1613	1540	807	**831**	**436**
5 h / 3 h / 0.46	173	1.80 h	1934	1149	967	574	**522**	**310**
1 h / 12 h / 0.46	73	0.39 h	2805	1470	1402	735	**757**	**396**
5 h / 12 h / 0.46	173	1.80 h	1369	813	684	406	**369**	**219**
1 h / 3 h / 0.3	73	0.39 h	4721	2474	2360	1237	**1275**	**668**
1 h / 3 h / 0.7	73	0.39 h	2023	1060	1011	530	**546**	**286**

Table 3: LNO$_x$ PE (mol NO$_x$ per flash) using different parameters and EUCLID lightning data.

Parameters Flash Window / Lifetime / AMFLNOX	Nflashes	Age	LNOx 10 th background - No corrected by DE	NOx 40 th background - No corrected by DE	**LNOx 10 th backgroud - Corrected by DE**	**LNOx 40 th backgroud - Corrected by DE**
1 h / 3 h / 0.46	812	0.33 h	276	155	**146**	**82**
5 h / 3 h / 0.46	1433	1.37 h	208	130	**89**	**56**
1 h / 12 h / 0.46	812	0.33 h	256	144	**136**	**76**
5 h / 12 h / 0.46	1433	1.37 h	161	101	**59**	**37**
1 h / 3 h / 0.3	812	0.33 h	424	238	**224**	**126**
1 h / 3 h / 0.7	812	0.33 h	176	102	**96**	**54**

Table 4: LNO_x PE (mol NO_x per flash) using different parameters and ENTLN lightning data.

12/07/2018:

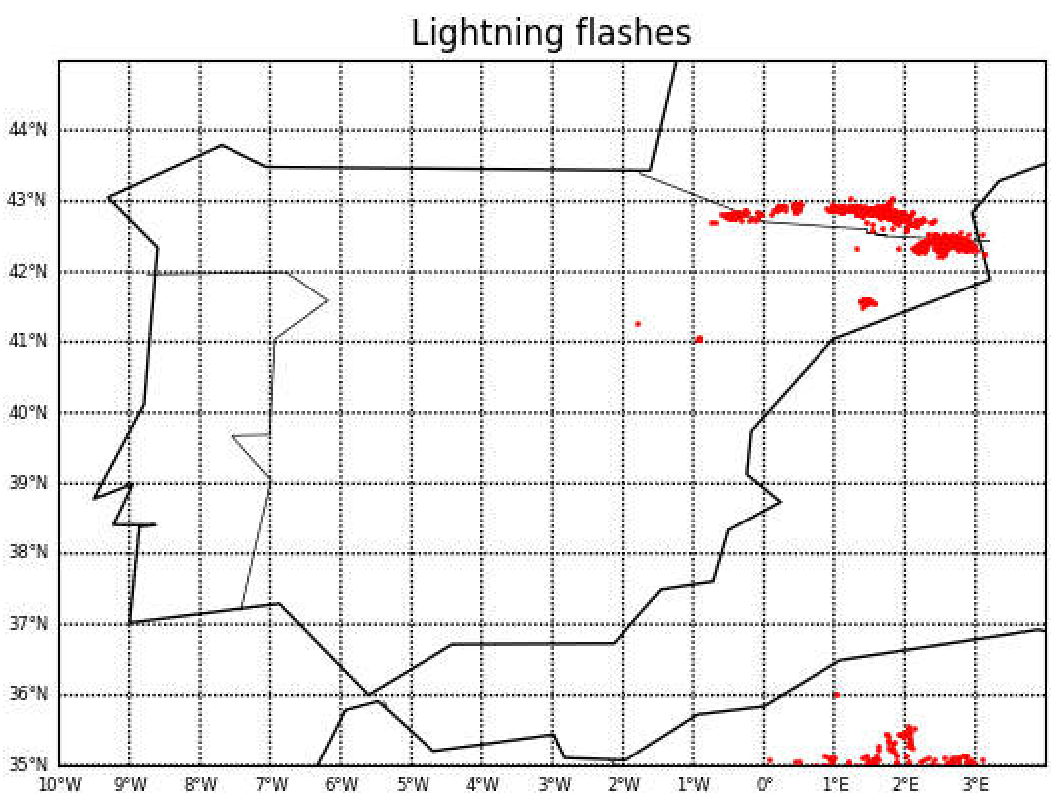

Figure 12: Lightning flashes provided by ENTLN 5 hours before the passage of TROPOMI.

CG/IC = 0.74

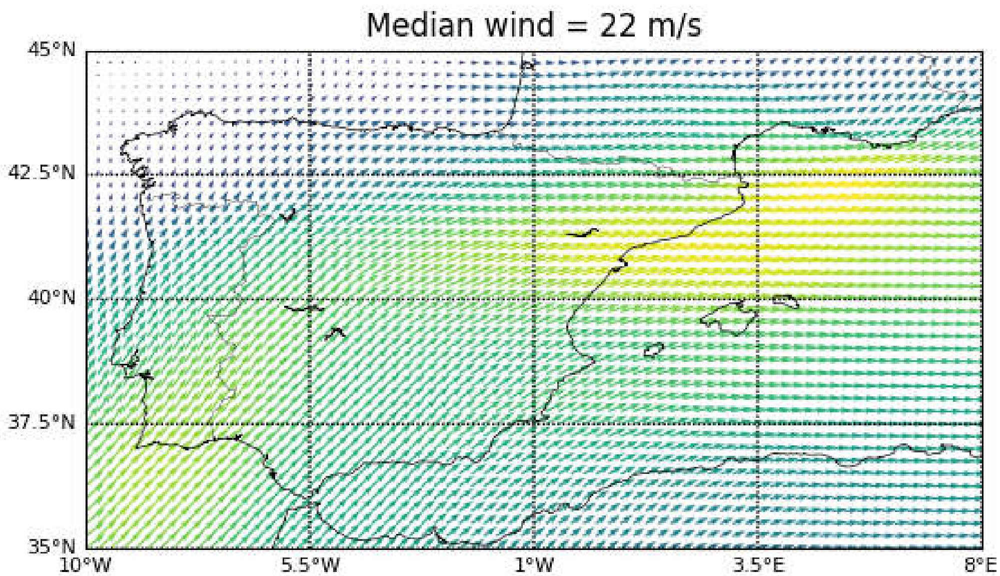

Figure 13: Median horizontal wind between 200 hPa and 500 hPa.

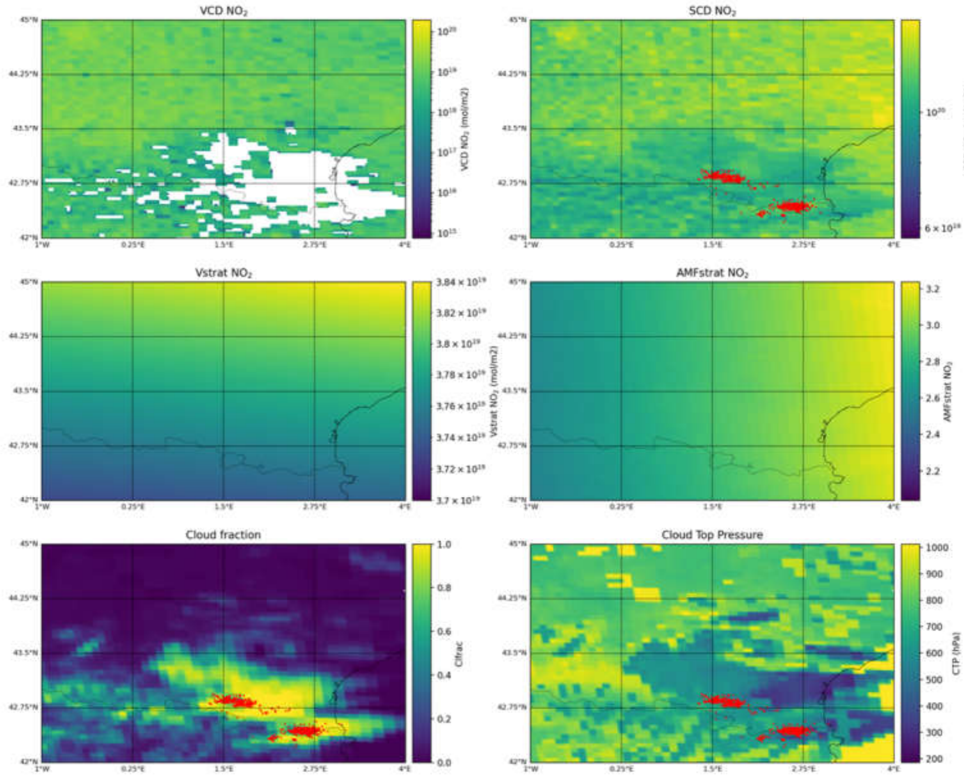

Figure 14: TROPOMI DLR-NO_2 product and flashes 1 hour before the passage of TROPOMI (red dots).

Parameters Flash Window / Lifetime / AMFLNOX	Nflashes	Age	LNOx 10 th background - No corrected by DE	NOx 40 th background - No corrected by DE	**LNOx 10 th backgroud - Corrected by DE**	**LNOx 40 th backgroud - Corrected by DE**
1 h / 3 h / 0.46	787	0.47 h	288	22	**164**	**13**
5 h / 3 h / 0.46	1143	1.19 h	233	148	**136**	**86**
1 h / 12 h / 0.46	787	0.47 h	,257	20	**147**	**11**
5 h / 12 h / 0.46	1143	1.19 h	187	119	**108**	**69**
1 h / 3 h / 0.3	787	0.47 h	441	34	**252**	**20**
1 h / 3 h / 0.7	787	0.47 h	189	15	**108**	**8**

Table 5: LNO_x PE (mol NO_x per flash) using different parameters and ENTLN lightning data.

DISCUSSION AND FUTURE WORK

Discussion

According to our results, LNO_x PE ranges between **88 and 756 mol NO_x per flash** using ENTLN lightning flash data, and between **345 and 2703 mol NO_x per flash** using EUCLID lightning data. In this section, we choose the LNO_x PE calculated using *DE = 0.27* because is less influenced by the clustering algorithm that EUCLID employs to cluster strokes into flashes. The following table shows the main sources of uncertainties in LNO_x:

Uncertainties

Parameter	Yielded LNOx PE differences
Flash window (1 – 5 h)	81%
NO_2 lifetime (3 – 12 h)	13%
Background estimation (10th – 40th %) ***without wind velocity correction***	542%
Background estimation (10th – 40th %) ***with wind velocity correction***	344%
AMFLNOx PE (0.3 – 0.7)	300%
LLS (EUCLID or ENTLN)	356%
NO_2 product over DCC pixels	?

Table 6: Uncertainties.

There are three different types of uncertainties in the obtained LNO_x PE:

1) LNO_x PE algorithm: Flash windows, NO_2 lifetime, background estimation, AMFLNOx and DCC pixels definition.

The most important sources of uncertainty associated with the employed LNO_x PE algorithm are the AMFLNOx and the background estimation. The definition of the DCC pixels could also introduce uncertainty in the final results.

We will use the numerical global atmosphere-chemistry model ***EMAC*** (ECHAM/MESSy Atmospheric Chemistry) to calculate the AMFLNOx for the investigated cases. We will also explore the possibility of using aircraft measurements or EMAC simulation to get a better estimation of the background NO_2.

The use of ERA5 wind velocities to discard cells influenced by LNO_x from background contributes to reduce the variability in the results (from 542% to 344%).

2) Lightning Location System Detection Efficiency.

The DE of Lightning Location Systems (LLS) over the Iberian Peninsula is relatively low. The launch of the *Meteosat Third Generation* (MTG) geostationary satellites of EUMETSAT in 2022 will provide a continuous monitoring of the occurrence of lightning flashes over the Iberian Peninsula through the instrument ***Lightning Imager*** (LI) from 2023 with a DE similar to the American Geostationary Lightning Mapper (~0.75). MTG-LI will significantly contribute to enhance our estimation of LNO_x over this region.

3) TROPOMI NO_2 product over deep convective systems.

Finally, the stratospheric NO_2 term and the fact that the NO_2 of the lower portion of the cloud is not visible for TROPOMI can be a significant source of uncertainty. We will quantify the influence of these terms in a future work.

Future work

- Quantify the influence of the stratospheric NO_2 term and the fact that the NO_2 of the lower portion of the cloud is not visible for TROPOMI.
- Reduce the uncertainty associated with AMFLNOx using the scattering weights calculated by Bucsela et al., (2013) and EMAC cases simulations.
- Explore the possibility of using aircraft measurements or EMAC simulation to get a better estimation of the background NO_2.
- Include more cases in the analysis in order to identify which uncertainties are systematic and which can be reduced with more case studies.
- Compare the results with LNO_x PE calculated using other TROPOMI-NO_2 products.

SUMMARY

ENTLN

Parameters Flash Window / Lifetime / AMFLNOX	LNOx 10 th background - Corrected by DE	LNOx 40 th background - Corrected by DE
1 h / 3 h / 0.46	493	134
5 h / 3 h / 0.46	287	213
1 h / 12 h / 0.46	436	119
5 h / 12 h / 0.46	195	144
1 h / 3 h / 0.3	756	205
1 h / 3 h / 0.7	324	88

EUCLID

Parameters Flash Window / Lifetime / AMFLNOx PE	LNOx PE 10 th background - Corrected by DE = 0.27	LNOx PE 40 th background - Corrected by DE = 0.27
1 h / 3 h / 0.46	1762	526
5 h / 3 h / 0.46	951	705
1 h / 12 h / 0.46	1561	469
5 h / 12 h / 0.46	643	476
1 h / 3 h / 0.3	2703	806
1 h / 3 h / 0.7	1158	345

Table 7: Averaged LNO_x (mol NO_x per flash) using different parameters.

We have estimated the LNO_x per flash in three thunderstorms over the Pyrenees using TROPOMI DLR-NO_2 measurements, lightning data from ENTLN, EUCLID and LIS-ISS, and wind velocity data from ERA5 reanalysis.

According to our results, LNO_x ranges between **88 and 756 mol NO_x per flash** using ENTLN lightning data, and between **345 and 5360 mol NO_x per flash** using EUCLID lightning data.

*The use of **ERA5 wind velocities** to discard cells influenced by LNO_x from background contributes to reduce the variability in the* *results (from 542% to 344%).*

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ABSTRACT

Lightning discharges are one of the main sources of atmospheric NO_X, contributing to about 10% of NO_X emissions globally and playing an important role in the concentration of ozone and other chemical species in the upper troposphere. Lightning produces between 2 and 8 Tg N per year globally [Schumann and Huntrieser, 2007], which corresponds to 100-400 mol NO_X per flash. Recent studies suggest that the production of NO_X per flash could depend on the length of the lightning channel, the type of lightning discharge and/or other factors than can vary between different thunderstorms or regions. Despite significant advances achieved by aircraft campaigns and by the improvement of satellites during the last two decades, reducing the uncertainty in the production of NO_X by lightning and understanding the factors that influence the production in different thunderstorms is still a challenge.

The TROPOspheric Monitoring Instrument (*TROPOMI*) is orbiting the Earth from a near-polar, sun-synchronous orbit since October 2017. TROPOMI is equipped with four spectrometers that provide information about the vertical chemical composition of the troposphere with an unprecedented horizontal spatial resolution of about 3.5 x 5.5 km. In this work, we use the DLR-NO_2 research product and the DLR cloud operational product to estimate the production of NO_X per flash from the Iberian Peninsula. We for the first time ever use chemical measurements from TROPOMI with lightning radio measurements provided by the EUropean Cooperation for LIghtning Detection (EUCLID) and the Earth Network Total Lightning Network (ENTLN), together with lightning optical measurements provided by the space-based Lightning Imaging Sensor (LIS). The use of different lightning detection systems allows us to estimate the Detection Efficiency (DE) of each system and to reduce the uncertainty in the production of NO_X per flash associated with the inhomogeneous DE in the studied region.

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