A TROPOMI- and GLM-based Estimate of NO_x Production by Lightning over the U.S.

Dale Allen[1], Kenneth Pickering[1], Eric Bucsela[2], Jos van Geffen[3], Henk Eskes[3], Nickolay Krotkov[4], William Koshak[5], Pepijn Veefkind[3], and Jeff Lapierre[6]

[1]University of Maryland, [2]SRI International, [3]Royal Netherlands Meteorological Institute, [4]NASA Goddard Space Flight Center, [5]NASA Marshall Space Flight Center, [6]Earth Networks

Introduction

- Lightning produces NO because the extreme temperatures (>20000 K) in lightning channels dissociate molecular O_2 and molecular N_2, which then combine to form NO which quickly reacts with O_3 to form NO_2. Lightning is responsible for 10-15% of NO_x emissions globally. This is 2 – 8 Tg N a^{-1} (*Schumann and Huntrieser*, 2007) or 100 to 400 mol per flash. Much of the uncertainty stems from limited knowledge of lightning NO_x production per flash (LNO_x PE) or per unit flash length.

- Most LNO_x is injected into mid- and upper-troposphere where away from deep convection its lifetime is long relative to lower troposphere NO_x. NO_x in this region enhances the concentrations of upper tropospheric NO_y, OH, and O_3 & contributes to positive radiative forcing by O_3 and negative forcing by CH_4.

- We have previously used OMI NO_2 to obtain estimates of LNO_x production per flash over the Gulf of Mexico (*Pickering et al.*, 2016, *JGR*), in convective events during NASA's TC4 field program (*Bucsela et al.*, 2010, *JGR*), and over broad regions of the tropics (*Allen et al., 2019, JGR*) and midlatitudes (*Bucsela et al., 2019, JGR).* In the latter studies, we obtained PE values of 170 ± 100 mol flash and 180 ± 100 mol flash, respectively.

TROPOMI LNO_x PE Algorithm

$$PE = [V_{tropLNOx} \times \Sigma\, Area] / [N_A \times \Sigma\, (Flashes \times \exp(-t/\tau))]$$

PE ≡ LNO_x Production Efficiency (moles NO_x/flash)
$V_{tropLNOx}$ ≡ Median vertical column density (VCD) of LNO_x over good quality (qa_value > 0.50) or good/fair quality (qa_value > 0.16[1]) pixels within ROI[2] that satisfy the DCC[3].
Area ≡ Area of pixels within ROI that satisfy the DCC or have P < 500 hPa and undefined cloud-fractions
N_A ≡ Avogadro's Number
Flashes ≡ Number of GLM or ENLN flashes[4] within ROI during 5 hour period before TROPOMI overpass[5]
t ≡ Age of individual flashes at the time of the overpass
τ ≡ Lifetime of NO_x in near field of convection (2, 3 (best guess), or 12 hours)

[1] Fair quality pixels have retrievals issues including in many cases $AMF_{trop} / AMF_{geo} < 0.1$
[2] Region of interest (ROI) ≡ Latitude-longitude region encompassing deep convective system
[3] Deep convective constraint (DCC) ≡ Cloud fraction[6] > 0.95 and cloud pressure[7] < 500 hPa
[4] GLM DE assumed to equal 78%. ENTLN DE for CG (IC) flashes assumed to equal 100 (79%)
[5] Overpass time ≡ Time TROPOMI exited ROI
[6] Cloud Fraction ≡ cloud_fraction_crb_nitrogendioxide_window variable from TROPOMI NO_2data
[7] Cloud pressure ≡ cloud_pressure_crb variable from TROPOMI support data

$V_{tropLNOx}$ = Median ($V_{tropNOx}$) – $V_{tropbkgn}$

$V_{tropNOx}$ = [S_{NO2} – avg ($V_{stratNO2} \times AMF_{strat}$)] / AMF_{LNOx} [avg over all pixels within ROI satisfying DCC]

S_{NO2} ≡ NO_2 Slant Column Density (SCD) for individual DCC pixels within ROI
$V_{stratNO2}$ ≡ Stratospheric VCD of NO_2 for DCC pixels within ROI
AMF_{strat} ≡ Stratospheric air mass factor for DCC pixels within ROI
AMF_{LNOx} ≡ AMF converting tropospheric slant column of NO_2 to vertical column of LNO_x.
$V_{tropbkgn}$ is estimated using 3 different methods
$V_{tropbkgn10}$ ($V_{tropbkgn40}$) ≡ 20th (40th) % of $V_{tropNOx}$ for non-flashing pixels within ROI satisfying DCC.
$V_{tropbcli}$ ≡ Mean value of $V_{tropNOx}$ for v1.3.x pixels satisfying the DCC within ROI on low-flash days between May 1 & Aug 22, 2019 (see Figure 1 below).

GOES-R GLM (*Goodman et al.*, 2013) is near-IR optical transient detector that detects lightning from changes in optical scene due to release of electromagnetic radiation
It is one of several instruments that are part of the payloads for NOAA's GOES-R series that includes GOES-16, operational at 75.2° W, and GOES-17, operational at 137.2°.

- Fig. 2 shows coverage region for GLM. GLM measures total number of CG & IC flashes with a spatial resolution of 8 km at nadir & 14 km at edge. Mean DE exceeds 70% but may be suppressed over inverted polarity storms, severe storms, and/or storms with deep liquid water path (*Koshak et al.*, 2018).
- This study uses flashes from GOES-16 GLM.

Fig. 1. $V_{tropbcli}$; Values obtained by applying 5° box car smoother to 1° x 1° gridded values of $V_{tropNOx}$ obtained using pixels on low-flash days (< 10000 GLM flashes in domain during 5-hour period preceding TROPOMI overpass) that are more than ~50 km distant from lightning.

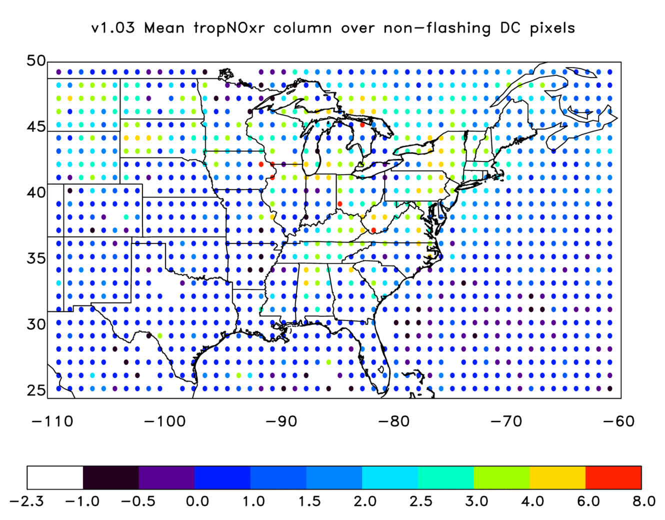
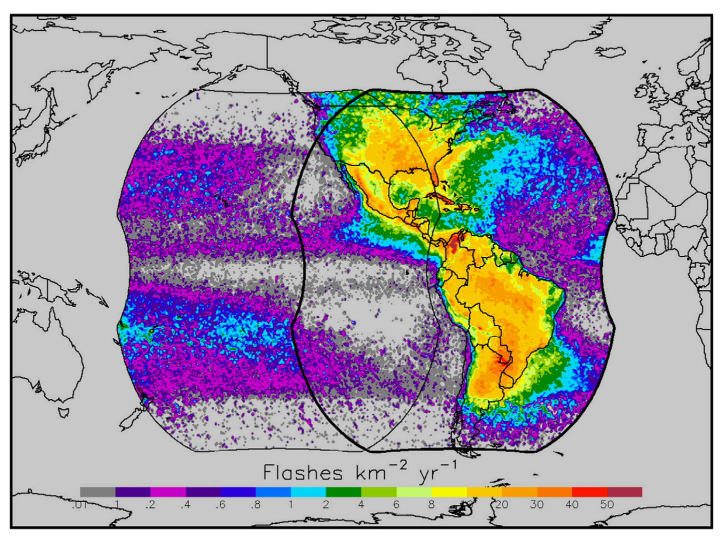


Fig. 2. Idealized representation of GLM flashes Observed by GLM-17 (left region) and GLM-16 (right region) from *Goodman et al.* [2013]



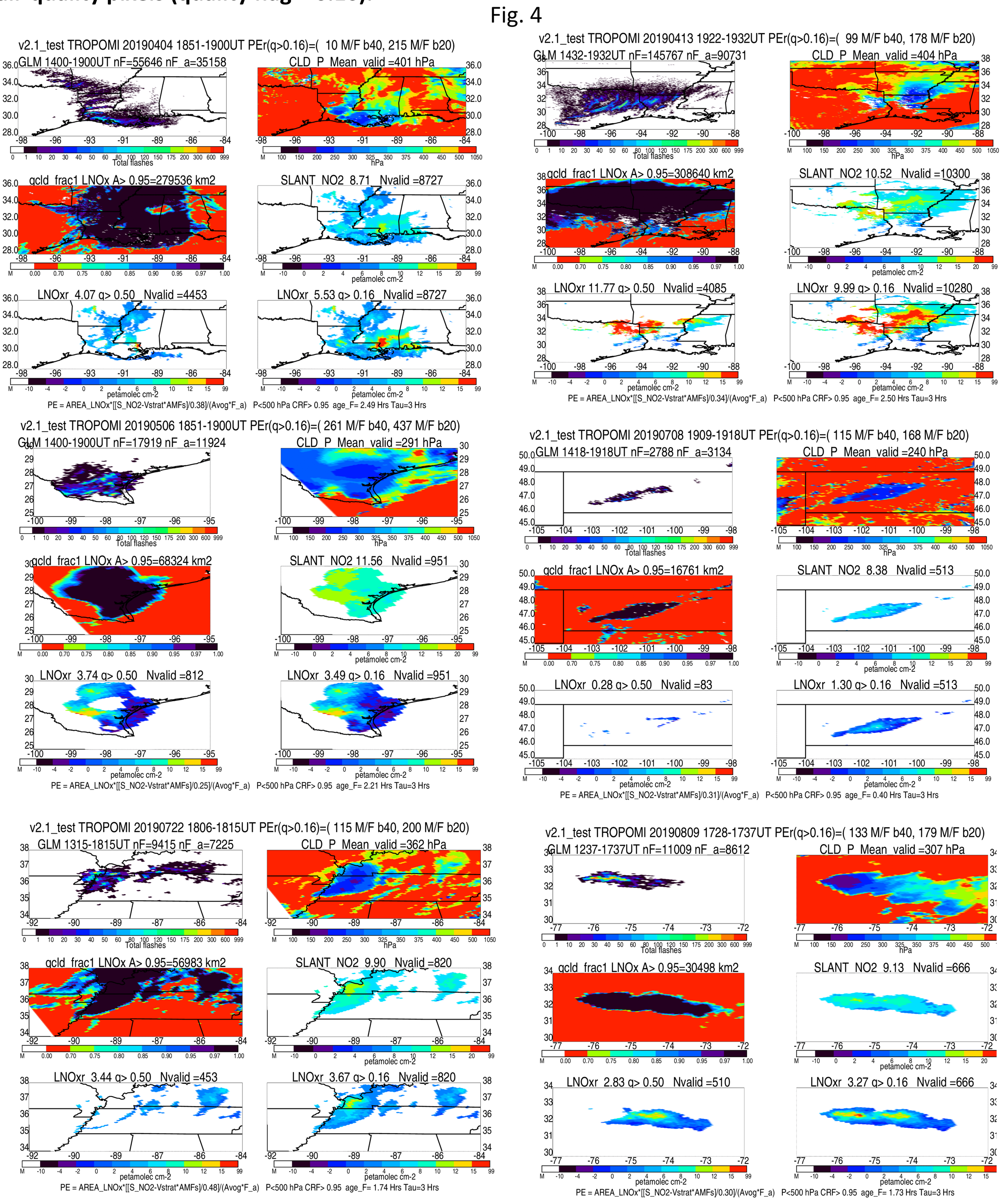
TROPOMI

Tropospheric Monitoring Instrument (TROPOMI) (*Veefkind et al.*, 2012) onboard the Copernicus Sentinel-5 Precursor satellite retrieves numerous trace gases including NO_2 and cloud products such as cloud fraction and cloud top pressure. The TROPOMI NO_2 processing system (*van Geffen et al.*, 2019, ATBD) is an improved version of the KNMI DOMINO system that retrieves 1) slant columns from Level 1b radiances using DOAS; 2) separates the tropospheric and stratospheric slant columns based on data from the TM5 model and assimilation system (*Huijnen et al.*, 2010); and 3) converts the tropospheric and stratospheric slant columns to vertical columns by application of air mass factors (AMF) which include daily information on NO_2 vertical distributions from the TM5 model at 1° x 1° resolution. The horizontal resolution of the NO_2 products at nadir are approximately 3.6 km (cross track) × 7.2 km (along-track) prior to August 6, 2019 and 3.6 × 5.6 km after August 6, 2019. The TROPOMI NO_2 retrieval uses cloud-pressures from the FRESCO-S algorithm, which is based on the FRESCO+ algorithm described in *Wang et al.* (2008). Cloud fraction information is retrieved from the NO_2 spectral window and accounts for Rayleigh scattering.

This study uses TROPOMI products from TROPOMI v1.01, v1.3.x (processor version 1.03), and v.2.1_test where the latter is a modified Copernicus Sentinel data product created for this study that includes spike removal to better deal with saturation and blooming effects in the radiance spectra allowing for increased data coverage over bright (flashing) scenes. Overexposure of CCDs (saturation) is common for TROPOMI scenes affected by lightning. Blooming occurs when the influence of saturation spreads to neighboring wavelengths and pixels.

Figures 4a-f show GLM and TROPOMI (v2.1_test) products over deep convection observed on April 4, 2019 (upper left), April 13, 2019 (upper right), May 6, 2019 (center-left), July 8, 2019 (center-right), July 22, 2019 (lower-left), and August 9, 2019 (lower right). For each day, the upper left panels show GLM flashes during the 5-hour period preceding the time of the TROPOMI overpass. nF (nF_a) gives the total flashes before (after) adjusting for chemical decay assuming a 3-hour lifetime. The upper right panels show the cloud pressure and also its mean over pixels satisfying the DCC. The mid-left panels show the cloud fraction in the NO_2 window and also give the area of pixels satisfying the DCC. The mid-right panels show S_{NO2} and give the number of DCC pixels for which S_{NO2} is defined. The lower-left panels show $V_{tropNOx}$ for good quality pixels (quality flag > 0.50). The mean value of $V_{tropNOx}$ and the number of pixels for which it is available are also shown. The lower-right panels also show $V_{tropNOx}$ but for good- and fair-quality pixels (quality flag > 0.16).

Fig. 4



YYYYMMDD	Region	Area	Nflashes	Age_Fl	Nflash_a	A_LNOx	VLNOxr	VLNOx_b40	VLNOx_b20	VLNOx_bkm	LNOxPEb40	LNOxPEb20	LNOxPEcli	NPTS
20180721c	86W-81W 28N-33N	81710	99561	2.71	43822	0.34	6.2	3.81	1.7	1.32	73	139	150	1715
20180723c	90W-82W 24N-30N	122176	37430	2.13	19944	0.33	11.77	10.69	7.11	0.25	109	473	1171	2738
20180726c	83W-79W 28N-31N	27874	40309	1.77	23947	0.36	3.01	3.1	2.26	0.23	-1	14	53	370
20190404c	98W-84W 28N-36N	279536	100829	2.48	49782	0.38	5.53	5.46	3.91	1.42	7	151	383	8727
20190413c	100W-88W 28N-38N	308640	225876	2.33	115778	0.34	9.99	8.23	6.84	1.03	77	139	396	10280
20190422c	98W-88W 38N-48N	22733	1083	2.86	430	0.62	1.24	0.84	-0.3	1.77	348	1347	-459	447
20190430c	105W-87W 33N-45N	287191	51190	1.91	30345	0.42	4.61	3.04	1.99	1.42	246	411	500	8101
20190505c	86W-78W 25N-31N	109140	90021	2.01	50770	0.33	4.43	0.78	-2.41	0.43	130	244	142	1733
20190506c1	102W-95W 38N-43N	49985	21858	1.7	13251	0.5	1.12	0.68	0.3	1.5	27	50	-24	1027
20190506c2	100W-95W 25N-30N	68324	42631	2.37	21087	0.25	3.49	0.74	-1.1	0.93	147	246	137	951
20190506c3	82W-73W 25N-31N	55903	21880	1.17	15552	0.47	-0.33	-1.46	-2.4	0.52	67	123	-50	1092
20190510c	98W-91W 25N-30N	60569	117907	2.74	52502	0.34	8.39	7.59	6.04	1.32	15	45	135	2173
20190528c	98W-88W 37N-43N	107665	56258	1.35	37303	0.52	1.67	0.77	-0.18	1.67	42	88	0	2578
20190531c	70W-60W 35N-40N	44355	53037	3.49	17637	0.53	3.65	2.01	0.92	1.09	68	114	106	845
20190608c	90W-78W 25N-35N	120395	110249	2.16	60204	0.36	2.7	2.39	1.32	1.15	10	45	51	3901
20190611c	87W-78W 25N-31N	45181	38536	1.36	25922	0.36	-0.18	-1.09	-2.8	0.44	26	75	-18	625
20190622c	102W-96W 40N-46N	31459	15192	2.62	6888	0.43	0.22	-0.18	-0.76	2.14	30	74	-145	999
20190623c	98W-88W 33N-38N	124840	102220	2.16	55823	0.29	1.9	2.03	0.95	0.97	-4	35	34	3070
20190630c	95W-87W 42N-48N	146174	155025	2.59	72248	0.5	2.71	1.75	0.3	1.83	32	81	29	3250
20190705c	99W-92W 36N-41N	88902	56078	2.31	28983	0.43	0.21	-0.29	-1.12	1.35	25	67	-58	1191
20190706c	88W-82W 27N-31N	74759	46214	1.89	26674	0.34	1.88	1.6	0.51	0.88	12	64	46	1038
20190708c	105W-98W 45N-50N	16761	13894	0.38	12280	0.31	1.3	0.01	-0.58	2.27	29	42	-21	513
20190713c	83W-77W 33N-36N	52490	75642	1.15	53282	0.4	1.96	0.94	-0.05	2.06	16	32	-1	788
20190716c1	96W-92W 41N-45N	19278	25152	2.55	11563	0.57	4.43	4.14	3.27	1.97	8	32	68	282
20190716c2	94W-88W 31N-36N	31520	15809	3.21	6180	0.44	3.31	0.57	-0.69	1.07	231	338	189	483
20190722c	92W-84W 34N-38N	56983	24726	1.69	14987	0.48	3.67	2.79	2.14	1.85	55	96	114	820
20190809c	77W-72W 30N-34N	30498	21612	1.7	13306	0.3	3.27	1	0.22	0.18	86	116	117	666
20190814c	92W-82W 28N-33N	82472	106588	1.46	69804	0.27	2.98	1.39	-0.15	1.64	31	61	26	3021
20190815c	100W-92W 38N-43N	56608	36756	2.59	17162	0.58	2.58	1.79	0.97	1.56	43	87	55	2191
All dates	All regions	89797	62192	2.1	33361	0.41	3.37	2.25	0.97	1.25	68	167	108	2262

Table 1. Details on the 29 case studies used to estimate LNO_x PE from GLM16 flashes and v2.1_test TROPOMI data. The table lists the date and location of each convective systems as well as details needed to estimate the LNO_x PE for each case.

In this table, area is given in km^2, the age of flashes (Age_Fl) is given in hours, VCDs of NO_x are given in peta molec cm^{-2}, and the PE is given in mol per flash. Nflashes gives the number of GLM flashes while Nflash_a is the number of flashes after adjusting for chemical decay assuming a chemical lifetime of 3 hours. Negative values of PE indicate that background columns over non-flashing grid boxes exceed the median columns in the region.

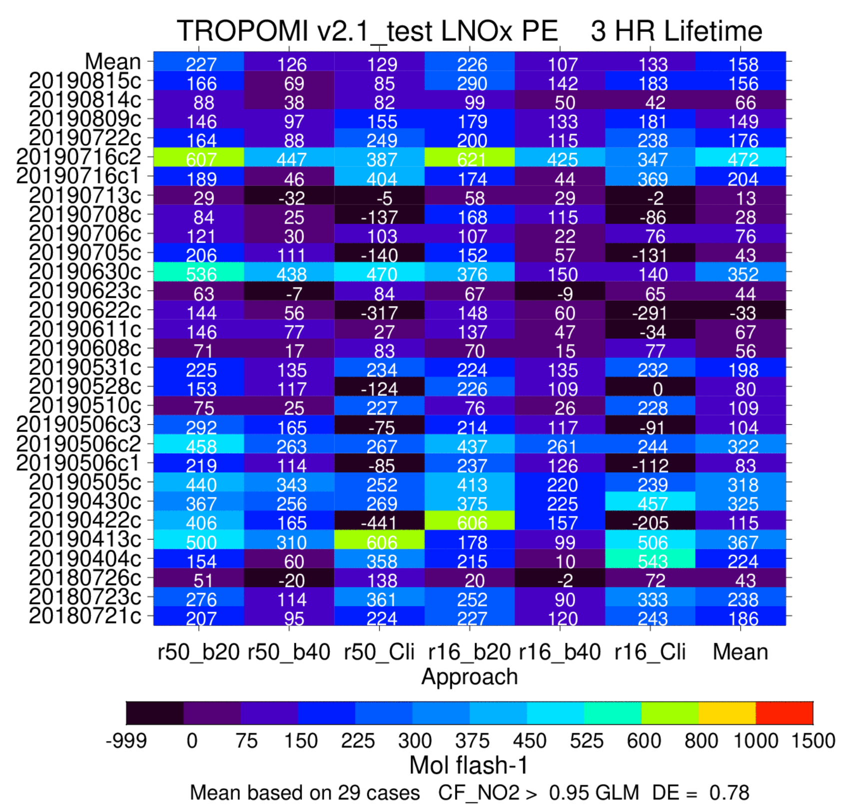


Table 2. LNOx PE assuming a 3-hour chemical lifetime for each of the 29 cases (y-axis) as derived using v2.1_test TROPOMI products and DE-adjusted GLM flashes for two TROPOMI quality flag thresholds (0.50 or 0.16) and three background assumptions (20%, 40%, or climatological).

The suffixes b20 and b40 indicate that the tropospheric background is day-specific and given by the 20th (40th)% column over non-flashing grid boxes within the footprint of the storm with CTPs < 500 hPa and cloud fractions > 0.95. The suffix Cli indicates that the mean background for flash-less deep-convective grid boxes over the summer of 2019 is used for that location.

The mean LNO_x PE over the 29 cases for the 6 approaches is 158 mol per flash. The mean PE for ENTLN flashes (not shown) is 112 mol per flash.

Tables 3 and 4 (below) show the mean LNO_x PE over the 29 cases as a function of lightning source (ENTLN or GLM), tropospheric background choice (climatological, b40, or b20), NO_x lifetime (2, 3, or 12 hours) and TROPOMI version (v1.3.x (left) and v2.1_test (right)). Colors show the standard deviations over the 29 cases.

LNO_x PE increases by more than a factor of two as assumed tropospheric background is decreased from 40 to 20%. Values for climatological background are in-between suggesting that the actual bkgn is between 20 and 40%.

LNO_x PE is ~50% greater for GLM flashes than ENTLN flashes suggesting that the assumed DE of 78% for GLM is too high or less likely that the assumed (100% for CG & 79% of IC flashes) DE for ENTLN is too low.

LNO_x PE decreases by approximately a factor of 2 as assumed lifetime varies between 2 and 12 hours.

LNO_x PE is ~20% higher for v2.1_test, which has fewer saturation issues, and consequently provides more robust estimates of LNO_x column and storm area.

LNOx PE for TROPOMI v1.3.x

	2	3	12
ENTLN_Cli	103	78	47
ENTLN_b40	80	60	36
ENTLN_b20	182	133	79
GLM_Cli	166	123	72
GLM_b40	129	96	58
GLM_b20	264	195	116

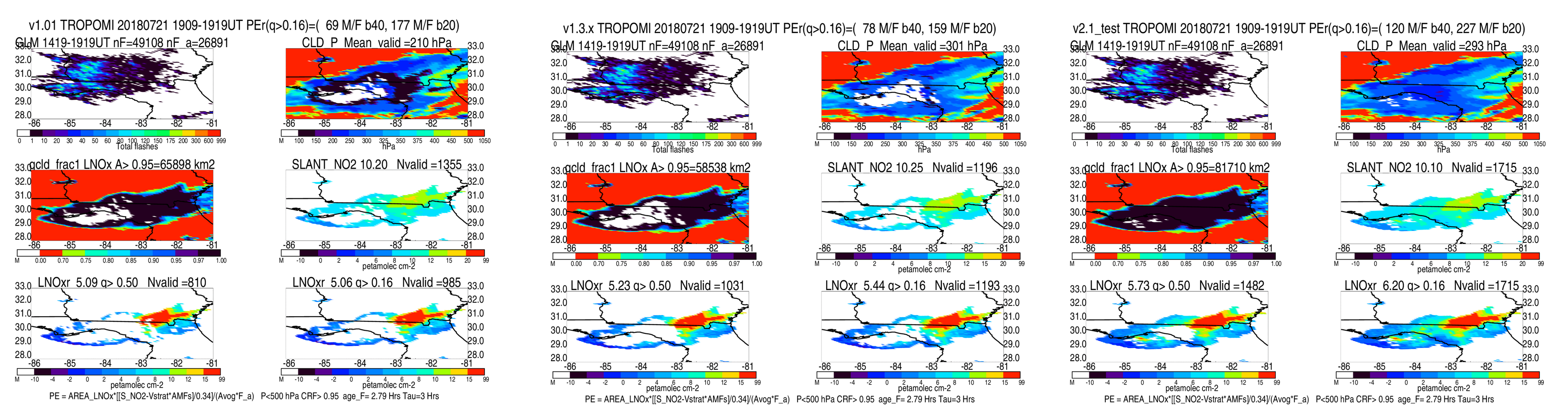
Lifetime
0 50 100 150 200 250 300 350 400
Standard deviation (Mol flash-1)
Mean and sigma based on 29 cases CF_NO2 > 0.95 qflag > 0.16

LNOx PE for TROPOMI v2.1_test

	2	3	12
ENTLN_Cli	138	108	69
ENTLN_b40	92	69	42
ENTLN_b20	228	167	100
GLM_Cli	177	133	79
GLM_b40	143	107	65
GLM_b20	307	226	135

Lifetime
0 50 100 150 200 250 300 350 400
Standard deviation (Mol flash-1)
Mean and sigma based on 29 cases CF_NO2 > 0.95 qflag > 0.16

Figures 3a-c show GLM and TROPOMI products over deep convection. TROPOMI products are shown for v1.01, v1.3.x, and v2.1_test. See Figure 4 caption for details on individual plots. For this system over the panhandle of Florida, the number of valid good (fair or good) quality VLNO$_x$ retrievals over pixels influenced by deep convection and/or lightning increased from 810 (985) in v1.01, to 1031 (1193) in v1.3.x, to 1482 (1715) in v2.1_test leading to more robust estimates of LNO_x PE.



Uncertainties

- AMFs used to convert SCDs of NO_2 to VCDs of NO_x vary with viewing geometry, Rayleigh and Mie scattering, the vertical profile of NO_2, and the NO / NO_2 ratio within a deep convective system (e.g., *Silvern et al.*, 2018).
- NO_x τ in near field of convection is assumed to equal 3 hours; it varies from 2-12 hours depending on proximity to deep convection (e.g., *Nault et al.* (2016).
- LNOx PE is sensitive to the VCD of NOx due to sources other than recent lightning (e.g., *Allen et al.*, 2019).
- DE for GLM flashes is assumed to equal 78%; which ignores storm-by-storm variations in DE. Comparison with ENTLN suggests this value is too high for these systems.
- TROPOMI columns are often missing due to saturation over bright regions where flashes and presumably VLNO$_x$ are large.
- Are these cases representative of deep convective systems over the United States and adjacent western Atlantic?

Summary

- LNO_x PE was estimated using GLM and ENTLN flashes and TROPOMI NO_2 columns for 29 convective systems observed during the spring- and summer of 2018-2019
- Mean LNO_x PE for a 3-hour lifetime ranged from 69 ± 83 mol per flash for ENTLN flashes and a 40% background to 226 ± 131 mol per flash for GLM flashes and a 20% background.
- Tropospheric NO_2 retrievals with TROPOMI are difficult over deep convective scenes due to small tropospheric AMFs, saturation of CCD pixels affected by lightning and blooming effects. However, tweaks to the processing algorithm allow more retrievals over these scenes.
- Future work will include refinement of the tropospheric background approach and analysis of the representativeness of these 29 cases.

- Acknowledgments: Much of this study is funded under the NASA Aura Science Team