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Estimating lightning NO_x production over South Africa

Nitrogen oxides ($NO_x = NO + NO_2$) are toxic air pollutants and play a significant role in tropospheric chemistry. Global NO_x hotspots are the industrialised regions of the USA, Europe, Middle East, East Asia and eastern parts of South Africa. Lightning is one of the many natural and anthropogenic sources of NO_x to the troposphere. It plays a role in the formation of particulate matter and tropospheric ozone, which are both linked to harmful health and climate effects. The discourse on NO_x over the southern African continent has mainly focused on anthropogenic sources. However, lightning is known to be a main source of tropospheric NO_x globally. It is therefore important to understand its contribution to the national and global NO_x budget. Data from the South African Lightning Detection Network were used to approximate the influence of lightning on the NO_x load over the country, and to develop a gridded data set of lightning-produced NO_x (LNO_x) emissions for the period 2008–2015. The Network monitors cloud-toground lightning strikes; and theoretically has a detection efficiency of 90% and a location accuracy of 0.5 km. An emission factor of 11.5 kg NO_2 /flash was employed to calculate the LNO_x budget of ~270 kt NO_2 /year. The calculated LNO_x was 14% of the total NO_x emission estimates published in the EDGAR v4.2 data set for the year 2008. The LNO_x emission inventory will improve model performance and prediction, and enhance the understanding of the contribution of lightning to ambient NO_2 .

Significance:

- The results show that both lightning and industrial NO_x sources are essential in evaluating NO_x and tropospheric O₃ chemistry over South Africa. As a result they should both be considered in air quality modelling, to assist in air quality management planning.
- LNO_x emissions are projected to increase with climate change, which may lead to an increase in tropospheric O₃. Thus it is important to have an LNO_x inventory, to be used as input into air quality modelling, as it will improve model performance and forecasting, and the understanding of the sensitivity of ambient pollution to changes in lightning emission.
- It will further inform chemical transport modelling so that the contribution of both natural and anthropogenic sources can be better understood.

Introduction

Poor air quality is a key environmental concern in South Africa, as it poses a serious threat to the well-being of the people of South Africa. Two of the key pollutants with adverse health and environmental impacts are nitrogen oxides (NO_x = nitric oxide (NO) + nitrogen dioxide (NO_2)) and particulate matter (PM). NO_x have effects that are felt on humans and the environment, but they are also important reagents in tropospheric chemical processes that result in the creation of secondary atmospheric pollutants, like ozone (O_3).

To alleviate the effects of air pollution, it is essential to have a comprehensive understanding of the sources of atmospheric pollutants to manage emissions. Management of air quality requires that the quantity of pollutants released into the atmosphere from sources be known, to determine how much of the emissions need to be reduced to achieve acceptable levels. Furthermore, the understanding of pollutant sources is essential for atmospheric chemical transport modelling, which is a valuable tool in comprehending the distribution of the pollutants and their potential impacts.¹

Areas that are strongly affected by poor air quality in South Africa include the Mpumalanga Highveld, Vaal Triangle area, and the Waterberg Bojanala area. These areas have been affirmed as air quality priority areas in terms of Section 19 of the *National Environmental Management: Air Quality Act.*² The primary sources of pollution in these areas are coal-fired power plants, vehicles, domestic fuel burning, and metallurgical and petrochemical industries.^{3,4}

Carbon monoxide (CO), ozone (O_3) , particulate matter (PM₁₀ and PM_{2.5}), nitrogen dioxide (NO₂) and sulfur dioxide (SO₂) are pollutants that are listed as being of particular concern in South Africa.⁵ These criteria pollutants are regulated by the National Ambient Air Quality Standards, as they have adverse health and environmental impacts.⁶ Nitrogen oxides are a focus of this study and are important in many ways:

- Long-term exposure to NO₂ may reduce the functionality of the lungs.⁷
- NO, increases the risk for children and the elderly of acquiring diseases such as bronchitis.8
- NO_x is vital in climate and atmospheric chemistry; it reacts with volatile organic compounds in the presence of sunlight to form O_x⁹ which in the troposphere has adverse health impacts and is a greenhouse gas¹⁰.

• NO_x is a precursor for the formation of secondary atmospheric aerosol, and it is involved in the formation of pollutants implicated in acid deposition.¹¹

Satellite analysis of total NO₂ vertical column density has identified some NO_x hotspots over the industrialised areas of the USA, Europe, Middle East, East Asia and South Africa (Figure 1).¹² South Africa's hotspot is over the northeastern parts of the country (the Highveld). This area contributes 90% of the industrial NO_x emissions (Figure 2).¹³ The Mpumalanga Province of South Africa is one of the world's largest NO₂ hotspots, with 12 coal-fired power plants situated in the area.¹⁴ Other sources of NO_x in the Highveld region include petrochemical industries (particularly coal and gas to liquid facilities), metallurgical smelters, road transport, biomass burning, and human settlements.¹³ Approximately 8 million people in the Highveld priority area and neighbouring Gauteng conurbation are exposed to poor air quality.¹⁴

Nitrogen oxides are released into the atmosphere from anthropogenic and natural sources. Anthropogenic sources are industrial activities, fossil fuel combustion, transportation and power plants. Natural sources include lightning and soil.¹⁵ Globally, 60–70% of the total NO_x budget is from anthropogenic sources rather than natural sources.¹⁶ Therefore, natural sources of NO_x in general, and lightning in particular, have received less attention. This is mainly due to the high spatiotemporal variability associated with the detection of lightning.¹⁷ Whilst this remains the case, lightning-produced NO_x (LNO_x) comprises about 10–15% of the global NO_x budget and is a leading source of NO_x in the upper troposphere.¹⁸ Consideration of the dynamics of LNO_x production is essential in understanding atmospheric chemical processes and the ambient concentrations of NO_x and O₃.¹⁹ Tropospheric O_3 concentration depends on the imminent precursor emissions, together with the changes in meteorological variables, including temperature and atmospheric moisture.²⁰ Various studies have suggested that future lightning activity is projected to intensify due to climate change. At the upper troposphere, LNO_x is more efficient in producing tropospheric O_3 ; thus, developing a regional LNO_x emission inventory will assist in understanding the sensitivity of the increase in lightning activity, with climate change, to O_3 in the troposphere.²¹⁻²³

During a thunderstorm, LNO_x is carried to the upper troposphere by convective updrafts, where it is more effective at generating O₃ and has a longer lifespan than in the lower troposphere, where most of the anthropogenic NO_x is released.²⁴ Thus, to be able to provide a reliable tropospheric O₃ budget, determination of an accurate LNO_x load is vital.¹⁹ Estimation of LNO_x is a challenge compared to that of anthropogenic emissions because the occurrence of lightning varies significantly in space and time.¹⁷ LNO_x production estimates range within the [1–20] Tg (N) per year, and most estimates from different studies point toward the lower end of the production range of ~ 5 Tg (N)/year.^{25,26} Although the production points towards the lower end, the evidence is not enough to reject the upper end.^{19,27,28}

In estimating LNO_x , a bottom-up approach is commonly used to measure (1) NO_x production per energy unit, (2) energy discharged per flash and (3) flash frequency, and to estimate LNO_x as the product of these quantities.¹⁷ Values cited in the literature range in magnitude, due to various assumptions and laboratory measurements. Another complication arises from the differences in cloud-to-ground (CG) and intra-cloud (IC) flashes. Previous studies suggested that IC flashes are more recurrent but less energetic than CG flashes (30% of total lightning), therefore less effective as NO_x producers compared to CG flashes (IC produces 10% of the NO_y produced by CG per flash).^{17,25,29}



Figure 1: Tropospheric NO₂ column density in 10¹⁵ molecules/cm² for March 2018.¹²



Figure 2: Tropospheric vertical NO, column over the Highveld of South Africa.¹³

However, recent studies have proposed that IC flashes may dissipate similar energy to that of CG flashes, and thus IC flashes may produce the same amount of NO_x as CG flashes.^{24,30-33} This is contradictory to the common assumption that IC flashes produce less NO_x than CG flashes, and highlights that, despite research efforts, there is still a great deal of uncertainty that remains regarding NO_x production on a per flash basis, as well as the relative production by IC and CG flashes. Additionally, calculations for LNO_x do not consider the effect that the water vapour has on the production of NO_x. Peyrous and Lapeyre (1982) have shown that NO_x production is strongly dependent on relative humidity.²⁵

There is currently limited research regarding the role of the production of NO_x by lightning in regional atmospheric chemistry. Of the studies that exist, most concentrate on the global LNO_x production.^{23,34,35} The global studies of LNO_x might not be applicable at the regional scale, because they may not capture the number of lightning strikes occurring within a region, the variability involving the production of NO_x per flash, and the varying ratio of IC and CG flashes.

This study builds on the work that has been done by Ojelede et al.³⁶ They conducted a study to estimate LNO_x production over the Highveld of South Africa for the year 2002, utilising lightning data obtained from the Lightning Position and Tracking System (LPATS) network. The LPATS has a detection efficiency of 80% and consists of six sensors over the eastern half of the country. The study was done on a limited spatial extent, for a short study period.

In this study, we used lightning data from the South African Lightning Detection Network (SALDN) that is operated by the South African Weather Service. It has coverage over the whole country, with a 90% detection efficiency for CG lightning and location accuracy of 0.5 km, i.e. the SALDN can detect at least 90% of all CG flashes and position them within 0.5 km. Information from SALDN allows for the high-resolution identification of areas of intense lightning occurrence. It also provides an opportunity to investigate the spatiotemporal characteristics of LNO_x production, which can then be used as an input into atmospheric chemical transport modelling and to improve air quality modelling. The data can also be used for validation of model simulations.

Data and methods

Lightning data from the SALDN were used in this research. The South African Weather Service installed the SALDN in 2005. Initially, it consisted of 19 sensors across South Africa but has since been expanded to 25 sensors including the one in eSwatini (Figure 3). The data are available from 2006 until the present; however, for this study, 8 years of data were used (2008–2015). The SALDN has a 90% detection efficiency of CG lightning and location accuracy of 0.5 km.³⁷ There have been some changes since the sensors were operationalised in 2006. Sensors have been added, and others have been relocated to ensure optimal network performance and to overcome some environmental developments and noise sources near some of the sensors. The network expansion and relocation of some sensors in 2009/2010, 2011, as well as in 2015, have greatly enhanced the accuracy of the lightning detection network; the accuracy is based on the South African Weather Service in-house quality control software.

The South African Weather Service network uses Vaisala instruments including two types of sensors: LS7000 and LS7001 model sensors. The LS7000 sensors were installed prior to 2009, and the LS7001 sensors were installed from 2009 onwards. These sensors measure the electromagnetic signature of a lightning flash using a low-frequency bandwidth because the radiation from CG strokes is strongest within that band.³⁸ As only a small percentage of IC flashes are measured at that bandwidth, the network sensors are primarily used to locate CG flashes.³⁸

The methods used to determine the position of individual CG strokes are (1) magnetic direction finding (MDF) and (2) time of arrival (TOA).³⁹ The combination of MDF and TOA techniques offers high detection efficiency and accurate location detection for CG lightning strokes from only two sensors. When the MDF and TOA methods are used individually, they require three or more sensors to detect lightning. It is impossible to get a 100% accurate position of lightning ground stroke even with combined MDF and TOA techniques. However, a degree of accuracy can be determined. For a thorough description of the method see Gill³⁹.

The lightning data retrieved using the Fault Analysis and Lightning Location System, contain information such as date and time, latitude,



Figure 3: Position of the 25 Vaisala lightning detection sensors over South Africa

longitude, and peak ampere. All CG lightning flash data were taken into consideration, regardless of polarity. Annual lightning ground flash density was calculated on $0.1^{\circ} \times 0.1^{\circ}$ grid boxes over the country and is expressed in flashes/km². The number of lightning flashes that were recorded by the SALDN over the 8 years (2008-2015) was counted for each individual grid box over the country. The sum of lightning flashes was divided by the area of the grid box to give the number of lightning flashes per square kilometre. To get the average annual lightning flash, the number of lightning flashes per square kilometre was divided by 8, as 8 years of data were considered. All grid boxes were considered, irrespective of the number of flashes. The Grid Analysis and Display System was used to display the lightning ground flash density map.

Information on total flash rate and production per flash is needed to quantify LNO, production. NO, emission factors from the literature and CG lightning flash data from the SALDN were used. In South Africa, there have been no studies done to estimate the IC/CG ratio. Therefore, IC/CG ratio estimates from other countries were used for South Africa (Table 1). Lightning characteristics – the number of strokes per flash, channel length, and ratios of IC and CG flashes – can vary.³⁴ Furthermore, the IC/CG flash ratio varies intensely during the life cycle of a thunderstorm. Therefore, it is assumed that such a ratio is a rough estimate as it is influenced by many factors, which include the severity and phase of a storm. Many studies have proposed a global annual mean IC/CG flash ratio of approximately 2 to 4.40 We used a IC/CG flash ratio of 3 - the most often used ratio for the mid-latitude to sub-tropic regions.^{25,41}

Various emission factors from different studies have been used (Table 2). We used the emission factor from Schumann and Huntrieser⁴⁰

for this study. Beirle et al.³⁵ referred to it as being the currently bestaccepted emission factor. Using the emission factor from Schumann and Huntrieser⁴⁰, one should bear in mind that global LNO, production might not be entirely applicable to regional LNO, production. Schumann and Huntrieser⁴⁰ concluded that the most likely value of global LNO, production is 5 Tg N/year with an uncertainty of 2-8 Tg N/year. The global annual mean flash frequency is 44±5/second, and a mean production per flash is 250 moles NO/flash (which is approximately 11.5 kg NO₂/flash).

Based on previous studies^{24,32,33,47}, we assumed that IC flashes generate the same amount of NO, as CG flashes. Total LNO, (kt NO,/year) to total flashes was calculated using Equation 1:

Total
$$(LNO_{y}) = P(NO_{2})(IC) + P(NO_{2})(CG)$$
 Equation 1

where

- $P(NO_2)$ is the production of $NO_2 = 11.5 \text{ kg } NO_2/\text{flash}$
- $IC = 3 \times CG$ flashes
- CG = CG flashes

Annual LNO, maps were generated to be able to observe the distribution of LNO, over South Africa. Equation 1 was used to calculate the total LNO, on a $0.1^{\circ} \times 0.1^{\circ}$ resolution over the country. The lightning flash information from the SALDN, the emission factor from Schumann and Huntrieser⁴⁰, and the IC/CG ratio of 3 were used for calculations.

Table 1: Intra-cloud to cloud-to-ground lightning flash ratio from different literature

IC/CG flash number ratio	Reference	Location			
3.35 (2 to 6)	Prentice and Mackerras ⁴²	Brisbane, Australia			
3	Price et al. ²⁵	USA			
4.4±1	Mackerras et al. ⁴³	Brisbane, Australia			
3.5	Nesbitt et al.44	USA			
Over 100	Dye et al. ⁴⁵	Northeastern Colorado, USA			
2.4	Bond et al. ⁴⁶	Tucson, Arizona, USA			
2.8±1.4 (1 to 9)	Boccippio et al. ⁴¹	Continental USA			

IC, intra-cloud; CG, cloud-to-ground

Table 2: Estimates of LNO, production per flash from different studies

Production rate (kg NO _z /flash)	Method	Reference			
3.5 (1.2 to 10.6)	GOME and NLDN	Beirle et al.49			
51.2	Theoretical	Price et al. ²⁵			
16.4	3D cloud model, in-situ measurements, EULINOX	Fehr et al.50			
4.1 (1.5 to 11.0)	GOME and NLDN	Beirle et al. ⁵¹			
11.5 (1.5 to 30.5)	Review	Schumann and Huntrieser ⁴⁰			
9.2 to 23.0	STERAO-A, ONERA, NLDN	DeCaria et al. ³²			
23	3D cloud model	Ott et al. ³³			
4.6 to 11.5	ОМІ	Bucsela et al. ⁵²			
11.4 (1.5 to 30.5)	SCIAMACHY	Beirle et al. ³⁵			

The total LNO_x emission is expressed in kg NO₂/km²/year. A box plot was created using the R-software lattice package to observe how LNO_x production varies from month to month.⁴⁸ The procedure for calculating LNO_x is similar to the one described above, expressed in kt NO₂/month. To produce the LNO_x diurnal variation graph, we used hourly lightning flash data for the 8-year period. The calculations were made over the 24-h period, considering every hour, from 00 hour to 23 hour, e.g. lightning flash considered for 00 hour is the one greater or equal to 00 hour but less than 01 hour.

Results and discussions

Figure 4 indicates the annual ground flash density for the entire country, showing how the lightning flash density is distributed over South Africa. The results show that high flash densities occur over Mpumalanga, KwaZulu-Natal and Gauteng Provinces, with the highest flash density over the far northern parts of KwaZulu-Natal and the Mpumalanga escarpment. The flash density declines towards the west of the country, along the coast and towards the northeastern parts of the country. Over the mountainous region of Lesotho, flash densities of ≤ 5 flashes/km² were observed. Gijben³⁷ and Gill³⁹ presented similar results.

Lightning is associated with convective storm development; hence portions of the country that experience many convective storms – the central, eastern and northern parts of the country – record high numbers of lightning flashes. The western and southwestern parts of the country are almost without thunderstorms, and therefore experience less lightning.³⁹ This is due to the influence of topography in enhancing thunderstorm development, and therefore increasing lightning activity.⁵³

Various studies internationally have shown a positive relationship between topography and lightning.⁵⁴⁻⁵⁶ The decrease in lightning over the elevated areas of Lesotho (Figure 4) can be attributed to the reduction in the occurrence of lightning at altitudes above 2000 m.^{37,53}

Figure 5 shows the spatial distribution of LNO_x over South Africa for the 8-year period (2008–2015). Areas of high lightning flash density (Figure 4) are regions that incur high LNO_x production. Highly elevated areas have a high production of NO_x , particularly over the northeastern parts of the country, except over the elevated areas of Lesotho where lightning occurrence is reduced because of altitudes above 2000 m. The Highveld region of South Africa is one of the regions where lightning is frequent and therefore has a high LNO_x production rate. This is also a region which is heavily industrialised and is the source of a large proportion of anthropogenic NO_x production.⁵⁷ In previous studies, in which LNO_x was not considered, it was found that the leading producer of NO_x in the Highveld Priority Area was industrial sources (power generation, petrochemicals and metallurgical industries) which are responsible for almost 96% of NO_y emissions.^{58,59}

Production of NO_x by lightning differs seasonally, in accord with lightning distribution. High production of LNO_x occurs in summer (NDJ) and autumn (FMA). This high LNO_x production is associated with deep convective thunderstorms, which are accompanied by lightning. During spring, a moderate amount of lightning occurs, while in winter there is close to zero lightning occurrence as there is lack of moisture and the atmosphere is stable, hence production is at a minimum during winter (MJJ).



Figure 4: Average number of lightning ground flashes per km² per year over South Africa for the 8-year period (2008–2015).



Figure 5: Average number of total LNO_x in (kg $NO_y/km^2/year$), using the emission factor of Schumann and Huntrieser⁴⁰.



Figure 6: Monthly estimates of LNO_X over South Africa.

Table 3 shows the total LNO_x generated, accompanied by the standard deviation of the interannual variability. The total LNO_x produced is estimated to be ~ 270.85 (\pm 42.5) kt NO₂/year, with summer contributing 51%, autumn 35%, spring 12%, and winter 2% of the annual budget. Figure 6 shows the variation of lightning production of NO_x from month to month, with the box-and-whisker plot exhibiting the minimum and maximum LNO_x production values, as well as the interquartile range and median. The lowest LNO_x production is noticeable in winter months and the highest production in summer months. The median values depict that LNO_x production further indicates relatively high values during spring and autumn as well, specifically in November, March and October.

 Table 3:
 Seasonal and annual LNO_x production over South Africa excluding Lesotho and eSwatini

Season	Total LNO _x production (kt)				
Summer	137 (± 27)				
Autumn	94 (± 21)				
Winter	6 (± 3)				
Spring	34 (± 11)				
Total LNO _x	270.85 (kt NO ₂ /year)				

The seasonal LNO_x estimates presented depict a similar pattern to that of Ojelede et al.'s³⁶ with summer contributing more to LNO_x annual estimates, followed by autumn, spring, and then winter. The total annual LNO_x production obtained by Ojelede et al.³⁶ was 24% of that recorded in this study. The 65 kt NO_x/year obtained is for when

IC flashes are assumed to produce only 10% of NO_x produced by CG flashes. Not assuming equal production of IC and CG flashes can lead to underestimation of LNO_x.³⁶

The diurnal variation of LNO_x in Figure 7 shows low production of NO₂ in the early morning, with peaks starting from ~0900 local time, reaching a maximum in the late afternoon around ~1500 local time before decreasing. The late afternoon peak in LNO_x may be because of the surface being heated due to the incoming solar radiation during the day, which then induces convective activity locally. Collier et al.⁶⁰ indicated the peak of annual lightning activity to be around 1700 local time in Pretoria, South Africa. Confirmation of these diurnal emission cycles improves the efforts to model LNO_x emissions over the country. The diurnal variation of LNO_x production is similar for summer, autumn and spring with LNO_x reaching its peak in the late afternoon, except during winter (Figure 8). During winter (green), there is not much variation in LNO_x production; it remains more or less the same throughout the day, and it is lower than during other seasons.

The calculation of the annual LNO_x production allows us to compare this particular source of NO_x to the South African atmosphere with the better known and understood industrial and vehicular sources, in order to determine the importance of LNO_x in the South African NO_x budget. Figure 9 shows NO_x emission from lightning, together with NO₂ emission obtained from the Emission Database for Global Atmospheric Research (EDGAR v4.2) for South Africa, which includes anthropogenic and biomass burning emissions.

The EDGAR v4.2 data used in the study provide estimates of the global biomass burning and anthropogenic emissions. The existing data are for all countries, with emissions given per main source category, on a spatial resolution of $0.1^{\circ} \times 0.1^{\circ}$ grid over the globe. The database was obtained from the European Commission's Joint Research Centre data catalogue for the entire South Africa.⁶¹ According to EDGAR v4.2 data (Table 4), the largest anthropogenic source of NO_x is the power generation sector (1A1a) which accounts for 1078.7 kt of NO₂ per year.



 $\label{eq:Figure 7:} Figure \ 7: \qquad \mbox{Mean annual diurnal variation of LNO_X production over South Africa for the years 2008 to 2015.}$



Figure 8: Seasonal diurnal variation of LNO_X production over South Africa.



Figure 9: NO, emission from EDGAR v4.2 and lightning for the year 2008 over South Africa, in kt NO_/year.

Table 4:	NO_2	emission	from	EDGAR	v4.2	and	LNO_x	for	the	year	2008
	over	South Afr	ica								

Sectors (code)	NO _x production (kt NO ₂ /year)					
Public electricity and heat production (1A1a)	1078.7 (57.5%)					
Road transportation (1A3b)	364.3 (19.4%)					
Lightning NO _x	270.9 (14.4%)					
Manufacturing industries and construction (1A2)	163.7 (8.7%)					
Grassland fires (5C)	132.6 (7.1%)					
Residential and other sectors (1A4)	65.4 (3.5%)					
Manure in pasture/range/paddock (4D2)	17.1 (0.9%)					
Domestic aviation (1A3a)	14.2 (0.8%)					
Direct soil emissions (4D1)	12.5 (0.7%)					
Rail transportation (1A3c)	10.7 (0.6%)					
Agricultural waste burning (4F)	3.8 (0.2%)					
Other energy industries (1A1bc)	3.2 (0.2%)					
Production of metals (2C)	2.6 (0.1%)					
Production of pulp/paper/food/drink (2D)	2.1 (0.1%)					
Forest fires (5A)	1.2 (0.1%)					
Manure management (4B)	0.9 (0.05%)					
Savanna burning (4E)	0.8 (0.04%)					
Production of chemicals (2B)	0.7 (0.03%)					
Fugitive emissions from solid fuels (1B1)	0.7 (0.03%)					
Fugitive emissions from oil and gas (1B2)	0.1 (0.01%)					

Road transportation (1A3b) is the second largest emitter of NO_x, emitting 365.3 kt of NO₂ over South Africa, accounting for 19.4% of the total NO_x from EDGAR v4.2. Road travel dominates the transport sector, with the vehicle/ownership ratio in South Africa higher than the world average.⁶² This is due to the extensive travel distances between settlements and workplaces.⁶² Lightning NO_x production is the third largest source of NO_x to the atmosphere in South Africa and accounts for ~ 270 kt NO₂/year, which is 14% of the total NO_x emitted as estimated in EDGAR v4.2. This percentage indicates that lightning is a significant source of NO_x in the atmosphere and cannot be ignored in air quality management planning; it also points to the importance of including lightning NO_x in the NO_x budget of the country.

The manufacturing industries and construction sector (1A2) is the fourth highest emitter of NO_x at 163.7 kt/NO₂/year and accounts for 8.7% of the total NO_x. Grassland fires (5C) and residential and other sectors (1A4) account for 7% and 3.4%, respectively, of the total NO_x emitted annually. The results indicate that industry is the main source of NO_x and road transportation is the second highest emitter of NO_x. Lightning NO_x production is the third highest emitter of NO_x. Delmas et al.⁶³ deduced that, at a global scale, combustion of fossil fuels contributes ~50% and biomass burning ~20%, while lightning and microbial activity in soils contribute ~30% of total NO_x emission. A recent study indicated that biogenic NO_x production over the Highveld region amounted to 28.25 kt/year, which was 3.9% of the total for the region. In the Waterberg, biogenic emissions of NO_x amounted to 3.72 kt/year, which is 2.28% of the total NO_y budget in the area.⁶⁴

Conclusions

We investigated the distribution of lightning over South Africa using 8 years of lightning data obtained from the SALDN. These emission factors gave an estimate of the total annual LNO_x production of 270.85 (± 42.5) kt NO₂/year. The LNO_x estimation in this study builds on the work of Ojelede et al.³⁶, who used 1 year of lightning data to estimate LNO_x production over the Highveld of South Africa. In this study, LNO_x production for the entire South African region was estimated over the 8 years. The Highveld region is the highest contributor to the total LNO_x production in the country.

The summer season accounts for 51% of LNO_x production, while the autumn season accounts for 35%. A sharp decline was experienced in winter, after which LNO_x production steadily increased in spring when transitioning from winter to summer. The diurnal variation of



a similar pattern in which LNO, was highest in the late afternoon and decreased in the evening after 20:00 UTC. Winter was the only season during which there was no diurnal variation. The characterisation of both the seasonal and diurnal dynamics of LNO, production is essential for its use in atmospheric chemical transport modelling and provides suitable input information.

Lightning NO, production contributed 14% to the national NO, budget, making it the third highest source, followed by power generation (57.5%) and road transportation (19.4%). These results highlight the importance of including lightning NO, estimates in the NO, budget for South Africa in further air quality management planning.

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Competing interests

We have no competing interests to declare.

Authors' contributions

B.M. led the writing of the manuscript and calculated the lightning NOx production over South Africa. G.F. and R.B. supervised the study and made contributions that helped improve the original manuscript.

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