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NO_x ground concentration at a station at high altitude in relation to cloud-to-ground lightning flashes

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Concentrations of both nitrogen oxides (NO_x=NO+NO₂) observed at a station at high altitude (Puy de Dome (PDD), 1465 m) have been analyzed in relation with the cloud-to-ground (CG) lightning activity provided by the detection network Météorage. A first analysis of the time series of both types of data issued for 4 months of the 2003 summer shows a good correspondence between high values of NO_x and stormy conditions in the area. During this 4-month period, NO_x values exceeding 100 μg m⁻³ have been observed on 7 days and all of these days correspond to lightning activity in a 200×200 km² area around PDD. Thirty-three days of the period experience lightning activity in this area, and only 14 of these days (42%) correspond to NO_x peak values exceeding 50 μg m⁻³. The presence of lightning activity is a necessary condition to have very large peak values of NO_x at PDD, but this condition is not sufficient. Two days showed NO_x exceeding 300 μg m⁻³ which represents a value between 176 and 270 ppbv according to the respective proportion of NO₂ and NO. Six days are analyzed in detail, using other information about the convective system and the wind conditions. The data of urban stations close to PDD have also been considered in the analysis for evaluating the possible transport by a plume from a city area. These days include the higher NO_x cases, cases with high lightning activity, and cases with remote lightning activity. On each of these days, the location of the CG flashes, the time delays between lightning activity and NO_x increase, and the air flow seem consistent with an interpretation of NO_x produced by lightning. The higher values of NO_x correspond with close and stationary thunderstorms which produced large proportions of positive CG flashes with low wind conditions. However, the characteristic increase seems to depend on several parameters of the CG flashes: their rate, location, and polarity. By comparing with

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the values given by other authors for aircraft measurements at altitude, our observations indicate that large quantities of NO_x could be produced by the CG flash processes.

Keywords: NO_x ground concentration; Cloud-to-ground lightning

1. Introduction

NO_x (NO+NO₂) are reactive in the photochemistry of the atmosphere, especially in ozone chemistry. So in the stratosphere, it may act as a catalyst in the ozone destruction reactions and in the troposphere, it may be considered as a major component in the sources and sinks of the ozone budget (Jaeglé et al., 1998). In the upper troposphere, NO_x may originate from stratospheric intrusions, from aircraft emissions, from surface emissions (including anthropogenic sources and biomass burnings) convectively transported from the boundary layer (Brasseur et al., 1996), and from lightning production (Levy et al., 1996). As a matter of fact, it is well known that the high temperatures in and around the lightning channel can cause the dissociation of the major atmospheric constituents N₂ and O₂ and the formation of NO_x in the free troposphere (Borucki and Chameides, 1984). The NO_x generated by lightning (LNO_x) could fill the lack in the estimated values for the NO_x sources budget in the upper troposphere (Murphy et al., 1993; Levy et al., 1996).

The global annual yield of LNO_x from lightning represents the largest uncertainty in the global NO_x budget, with estimates varying between 2 and 20 Tg (N) (Levy et al., 1996; Lee et al., 1997). A recent study from satellite data estimated this production at 2.8 (0.8–14) Tg (N)/year (Beirle et al., 2004). During the summer of 1996 over Southern Germany and Switzerland, the first airborne NO_x measurements in anvils of active thunderstorms in Europe were performed. By comparing the measurements in several types of convective clouds (cumulus congestus and thunderclouds), Huntrieser et al. (1998) found that 60–75% of the total NO_x present in anvils could be due to lightning flashes. NO_x concentrations were found to range from 1 to 4 parts per billion by volume (ppbv) (Höller et al., 1999). The STERAO–Deep Convection Field Project with closely coordinated chemical, dynamical, electrical, and microphysical observations conducted in northeastern Colorado during June and July of 1996 investigated the production of NO_x by lightning (Dye et al., 2000; Skamarock et al., 2003). The combination of these different means indicated that most of the NO_x produced by the storm was transported within the anvil, 60% of which was produced by lightning. In this study and from one storm case, the authors noted that the NO_x produced by lightning was almost exclusively from IC flashes with a ratio of IC to total flashes >0.95 throughout most of the storm’s lifetime, and they argued that IC flashes can be major contributors to NO_x production probably for all storms. NO peak values up to 19 ppbv were observed in the storms observed during STERAO (Stith et al., 1999). From this experiment, by using the observation results and cloud model simulations, estimations of LNO_x produced per meter of flash channel were made (Skamarock et al., 2003). So, the rate was estimated to about 1.7×10^{-3} mol/meter of flash channel.

In order to reduce the major uncertainty in quantifying the role of lightning in NO_x production in the troposphere, it is necessary to obtain accurate statistics on the energy, the length, the duration, the frequency of lightning, and the associated LNO_x production. On the

other hand, the transport has to be better estimated to well understand the role of the NO_x in the atmosphere (Skamarock et al., 2003). In this paper, we develop an analysis of NO_x data measured at the ground at a high-altitude site, in relation with cloud-to-ground (CG) lightning activity and convective cloud system characteristics. This analysis is made globally over a 4-month period in 2003 spring and summer, and in detail for 6 days. So, it is possible to compare NO_x values observed at the ground with NO_x values observed at altitude by other authors.

2. Data

The CG lightning flash data are issued from the French lightning-detection network managed by the Météorage firm. This network now includes 17 sensors equipped with both Direction Finder (DF) and Time Of Arrival (TOA) techniques. More information about the detection and location combining DF technique and TOA technique can be found in Cummins et al. (1998). The efficiency and accuracy of the network are now characterized by an accuracy between 500 m and 2 km for 50% of the CG flashes detected (Montariol et al., 2000). The data given by this network allow characterizing the CG activity with the following parameters: the location of the ground impact of each CG flash, its occurrence time, its polarity, its number of strokes (multiplicity), and the peak current of the strokes.

The French territory is covered by the meteorological radar network ARAMIS (Application Radar À la Météorologie Infra-Synoptique) from Météo-France (Chêze, 1990). It consists of 18 conventional radars, of which 10 are C-band radars ($\lambda=5$ cm) and 8 are S-band radars ($\lambda=10$ cm). Data from one of these radars are used in this study in order to describe the cloud systems associated with the lightning activity in the study cases. It is an S-band radar located in Grêzes (45.1°N; 1.37°E). This radar is chosen for its wavelength in the S-band where the attenuation by the rain is lower and for its location at about 150 km from the Puy de Dôme in order to scan the convective systems around the site at a level high enough. Its location is indicated in Fig. 1 with the label RADAR. The radar has a range of approximately 250 km and produces images every 5 min. The parameter provided by the radar is the reflectivity factor directly related to the precipitation rate. The images are Plan Position Indicator type, with a constant and low elevation of, respectively, 1.4° (short distance) and 0.4° (long distance).

The NO_x data are issued from local measurements at a high-altitude site from the Auvergne AASQA (“Association Agréée de Surveillance de la Qualité de l’Air”—Approved Association for the Observation of the Air Quality): ATMOAUVERGNE (<http://www.atmoauvergne.asso.fr/>). The French territory is covered with such stations managed by an AASQA in each administrative region (<http://www.buldair.org/>). This site is located in Auvergne region at the summit of the Puy de Dome at an altitude of 1465 m (45.79°N; 2.96°E; Fig. 1). The station is well located as a reference for regional scale chemistry studies because it is out of the boundary layer in quiet conditions. Both constituents NO and NO_2 are analyzed with a Model 42C from Thermo Electron Corporation. The principle of this device is chemiluminescence and it is capable of measuring oxides of nitrogen from sub parts per billion (ppb) to 100 parts per million (ppm). The sampling time of this analyzer is about 1 min, what is well adapted for the average time of the available data. The NO_x data are recorded in 15-min averaged

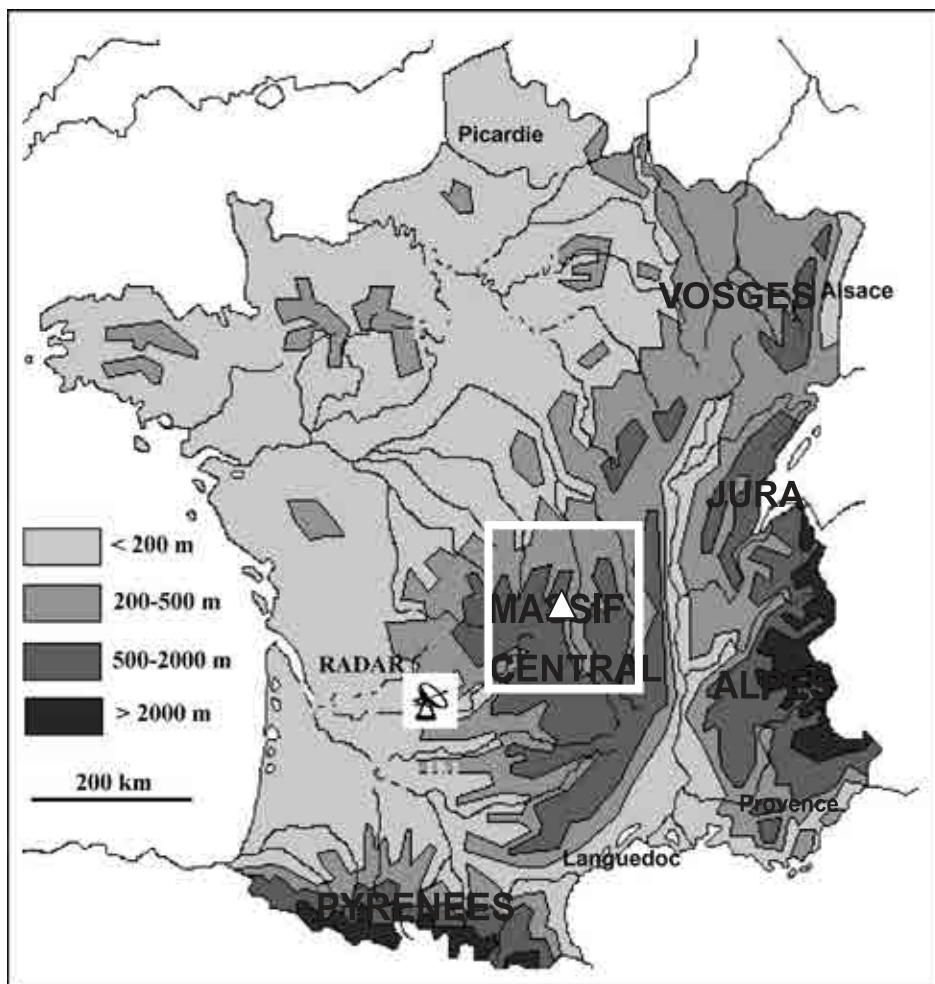


Fig. 1. Map of France with relief and with the locations of the radar of Grèzes (radar symbol) and of Puy de Dome station (white triangle) for chemistry measurements. The white frame indicates the $200 \times 200 \text{ km}^2$ area around PDD where the CG lightning activity is considered.

concentrations and a value represents the average over the past 15 min. In the following, PDD means the station of Puy de Dôme.

3. Data analysis

3.1. Overview

Fig. 2 displays the time series of NO_x and of the daily number of CG lightning flashes during May, June, July, and August of 2003. The CG flashes are counted in a 200×200

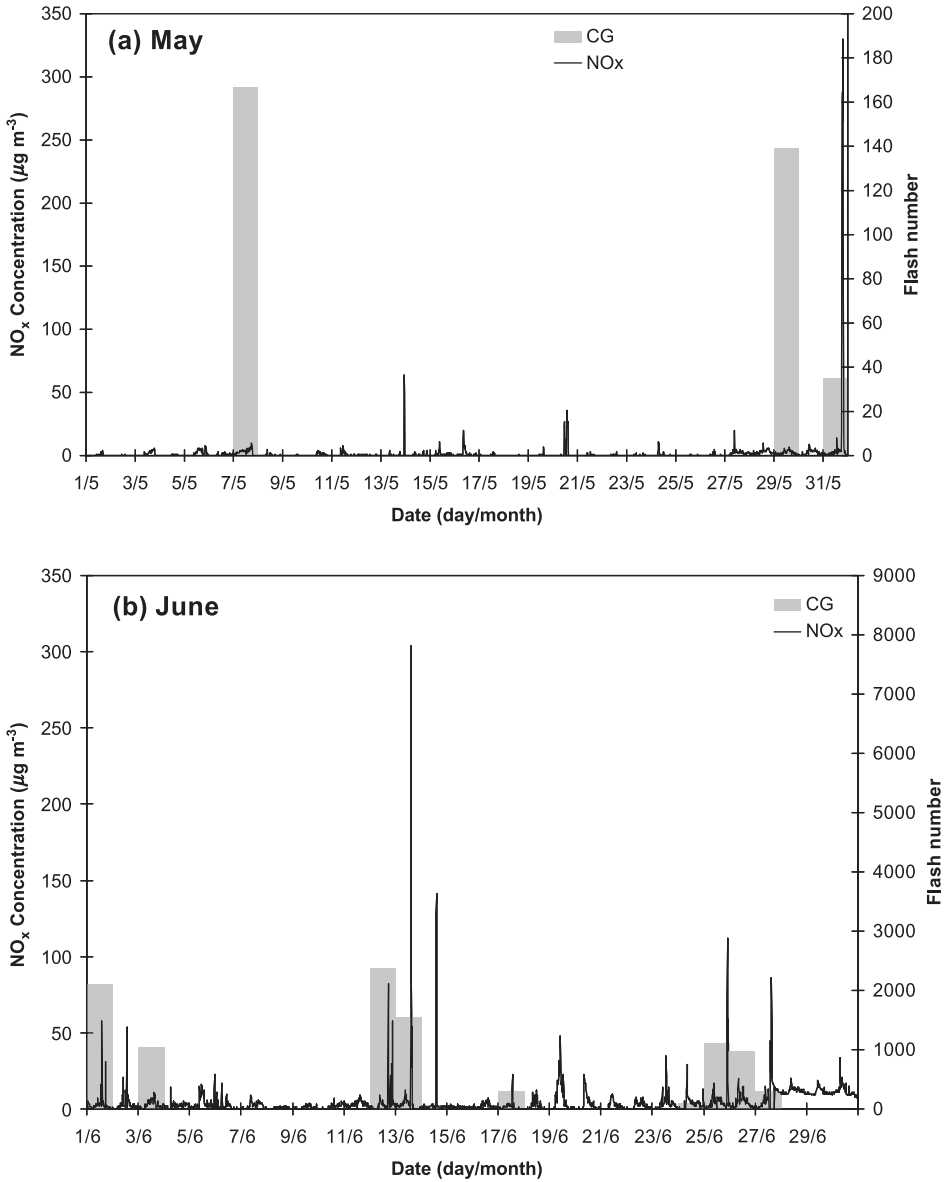


Fig. 2. Time series of the daily CG flash number (histogram), the 15-min NO_x measured at PDD (black curve) for (a) May, (b) June, (c) July, and (d) August. The CG flashes are counted in the 200×200 km² area. (e) Same for August but with the time series of NO₂.

km² frame centered on PDD (Fig. 1) and NO_x is considered as a 15-min average. The NO_x evolution displays a daily cycle most of the time with an increase during the day. It is especially visible in August (Fig. 2d) and the characteristic of this increase is a rise steeper than the declivity. During such increases, the values are generally of some µg m⁻³ and

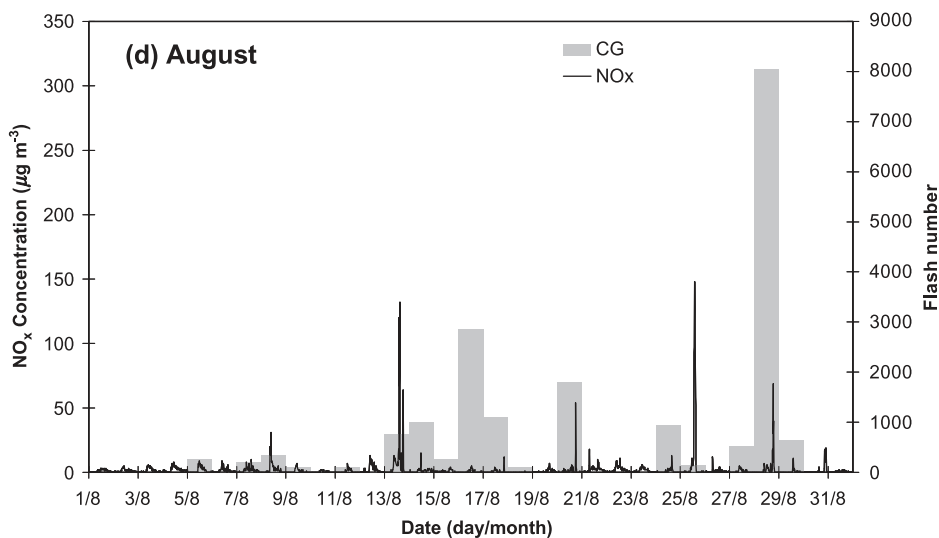
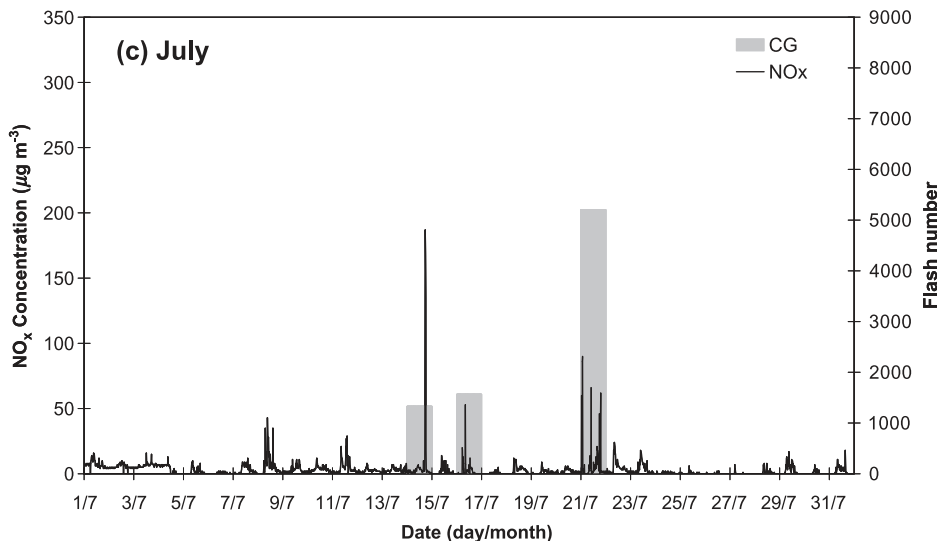


Fig. 2 (continued).

they correspond to days without any CG flashes in the area. The time series of NO_2 for the month of August (Fig. 2e) shows that these daily increases are mainly due to this constituent and therefore do not correspond to a local source of NO. In order to explain them, we may consider the arrival of polluted air masses from areas affected by human activity. Some days, NO_x undergoes large peaks, as for example on 31 May, 12, 13, 14, 26, and 27 June, 14, 16 and 21 July, 13, 20, 25, and 28 August. Some of these peaks largely exceed $100 \mu\text{g m}^{-3}$ and two exceed $300 \mu\text{g m}^{-3}$ on 31 May and on 13 June, $330 \mu\text{g m}^{-3}$

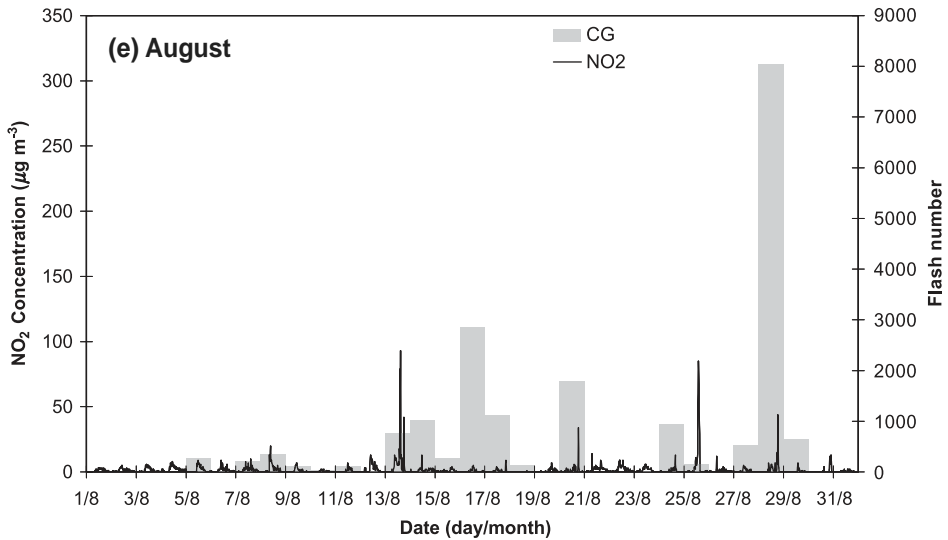


Fig. 2 (continued).

and $304 \mu\text{g m}^{-3}$, respectively. By considering the time series of the daily flash number, all events corresponding to the large peaks mentioned are days with lightning activity in the $200 \times 200 \text{ km}^2$ area. So, for example, the daily flash numbers for 21 July and 28 August are about 5206 and 8045, respectively. On the other hand, some days correspond to lightning activity without any apparent large increase in NO_x , for example, on 16 August. Furthermore, on this day, the CG flash number recorded for the area is high, 2849 CG flashes. The correspondence between NO_x peak values and the presence of lightning activity close to the site could be a first indication of the production of large amounts of LNO_x but it is necessary to make more detailed analysis to point out the exact mechanism responsible for the presence of NO_x at the measurement site. If the large NO_x increases observed at the PDD are influenced by CG lightning flashes, the amplitude of these increases may depend on several parameters related to the lightning flashes (the number, the rate, the location, the type) and on other parameters related to the meteorological conditions (the wind, the rainfall). Especially, we have to consider if the NO_x increase can be influenced by the polluted air from the city of Clermont–Ferrand located about 10 km and east of PDD. As it is difficult to consider a correlation study because of the large number of parameters, several days are analyzed in detail. So, in the next section, six case studies are developed in order to interpret the general observations presented in this section.

3.2. Case studies

Several detailed case studies are made to estimate the correlation between lightning activity and NO_x measured at PDD, especially to point out the chronology between the NO_x increase and the lightning location. The convective systems can be described with radar images. The wind direction can be determined from an on-line trajectory calculation model (HYSPLIT On-line Transport and Dispersion Model) (Draxler and Rolph, 2003).

Six days are considered in this study: some correspond to large NO_x peak values and various CG flash numbers and other correspond to CG activity without any NO_x peak value. Some days can display several periods of activity. Table 1 summarizes the characteristics of these days, by separating the different periods when it is necessary. The displacement speed of the system can be deduced from radar scans made every 5 min. The local wind characteristics (direction and velocity) can be provided from the meteorological station at PDD, available on the website of the observatory in Earth Physics at Clermont-Ferrand: (<http://www.obs.univ-bpclermont.fr/atmos/mesuresyst.htm>).

3.2.1. 31 May 2003

On 31 May 2003, the meteorological situation was characterized by a deep trough of low pressure located west of Ireland with a minimum of about 990 hPa at the ground. However, over France, the flow was weak and high temperatures became widespread. The CAPE value deduced from the sounding made in Bordeaux at 0000 UT on 31 May, was 400 J kg^{-1} . Fig. 3a displays the time series for the afternoon of both NO and NO_x , and of CG flash rates in the $200 \times 200 \text{ km}^2$ area, by distinguishing both polarities. The peak values of the CG lightning flash rates occurred at 1800 and 1815 UT, and they were 0.2 and 0.5 min^{-1} , for negative and positive CG, respectively. Two peaks of NO_x occurred: $288 \mu\text{g m}^{-3}$ at 1900 and $330 \mu\text{g m}^{-3}$ at 1930 UT. A large proportion of NO_x (about 50%) corresponded to NO, which would indicate the effect of a local source. The time delay between the first peak of CG flash rate and the first peak of NO_x was about 45 min. Therefore, in this case, the CG rate was low while the NO_x peak values were very high. However, we observed a good correspondence between both parameters since the CG flashes occurred first. The positive CG flashes drastically dominated, especially between 1800 and 1830 UT. For the whole system, 23 CG flashes were observed with only 5 negative ones. The fraction of positive CG flashes was therefore 79% for the whole lifetime, what is really untypical. The average peak currents were -9.8 and 42.9 kA for negative and positive flashes, respectively. The negative peak currents were therefore very low compared with typical values, for example, from the observations made by Orville et al. (2002) in the United States. All these observations seem to indicate a reversed dipole for this storm. All CG flash locations are indicated in Fig. 3b where PDD is located in (0;0). Most of them form a core at a few kilometers from PDD in the North-Westerly

Table 1
General characteristics of the thunderstorm events considered in the case studies

Day	CG flash number	Positive CG proportion (%)	Displacement speed (m s^{-1})	NO_x maximum ($\mu\text{g m}^{-3}$)
31 May	19	79	0.5	330
13 June	1547	36.5	1	304
21 July				
Period I, 00:00–05:00	2005	10	15	90
Period II, 05:00–12:00	1940	5	15	66
Period III, 12:00–20:00	1261	11	18	62
13 August	765	37	5	132
16 August	2849	14	10	5
28 August	8045	10	12	69

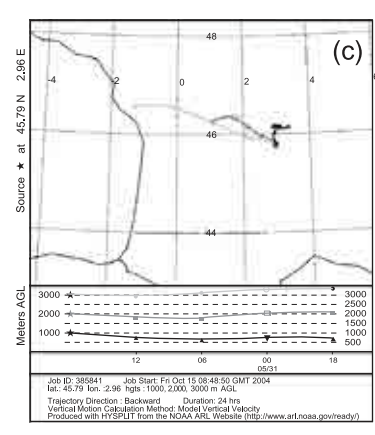
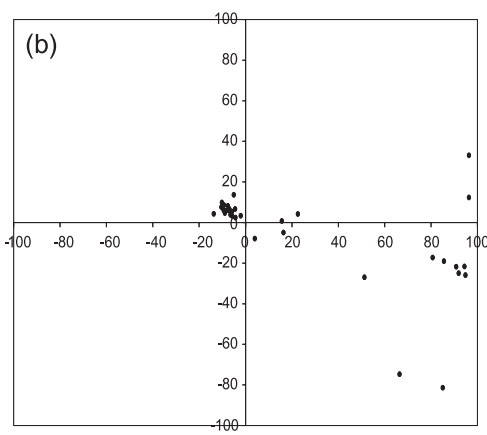
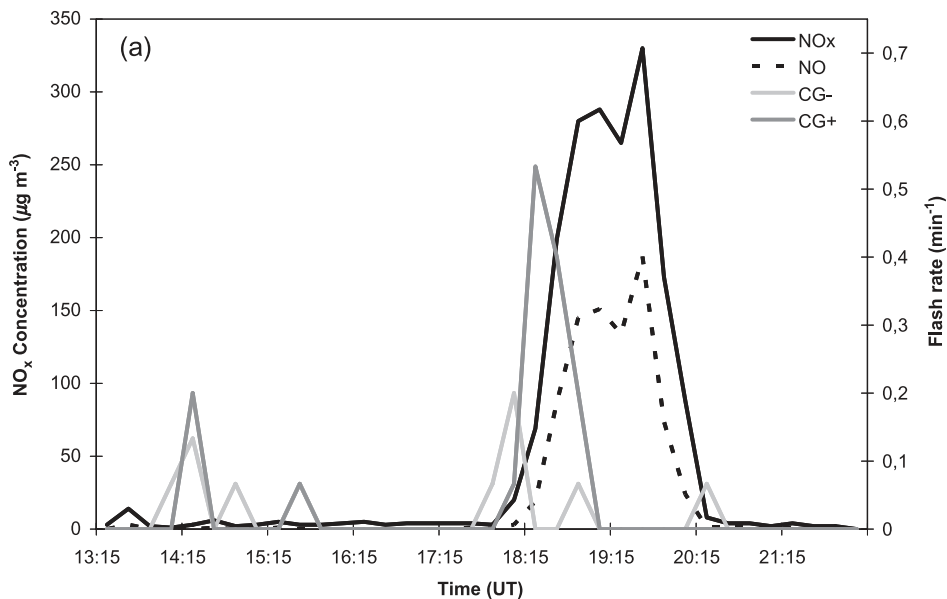


Fig. 3. 31 May: (a) Time series of NO_x and NO measured at PDD (black curves), negative and positive CG flash rates (grey curves). The time resolution is 15 min. (b) Diagram of the CG locations in the $200 \times 200 \text{ km}^2$ area. (c) Backward trajectories of the air motion from PDD calculated at three levels (1000, 2000, and 3000 m above ground). Latitude and longitude are in degrees.

direction. Fig. 3c displays the backward trajectory of the air motion calculated with HYSPLIT at several levels and for 24 h. These trajectories can support the transport of NO_x produced by the flashes up to PDD. At low level, at 1000 m above ground in this case, the velocity seems low since the trajectory is very short. The locally measured wind characteristics were not available for this day.

Fig. 4a and b display the radar scans of the system at 1800 and 1830 UT, respectively. The location of the CG flashes produced during 10 min around the

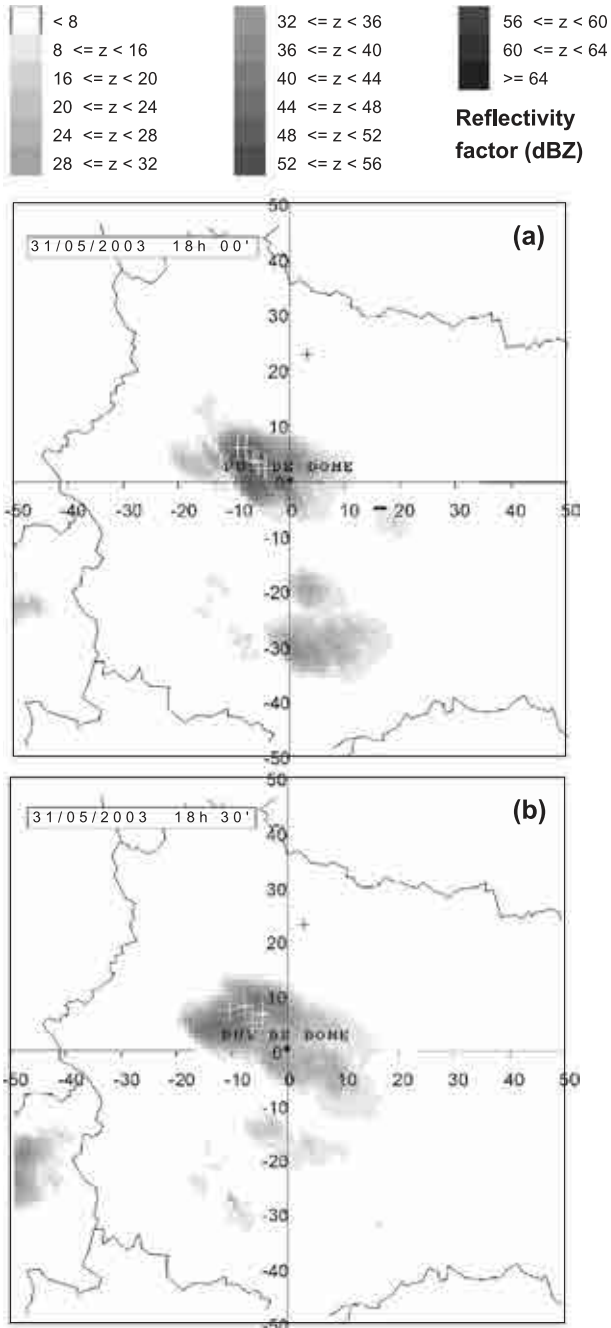


Fig. 4. Radar reflectivity horizontal field for 31 May in the $100 \times 100 \text{ km}^2$ area around Puy de Dôme: (a) at 1800 UT, (b) at 1830 UT. The reflectivity factor is in dBZ. The CG flashes detected during 10 min around the time of the radar scan are superimposed with white symbols (+) and (-) for positive and negative CG, respectively.

scanning times are superimposed with white symbols. We verify with both images that the system was really stationary and the CG flashes, characterized by a large majority of positive ones, were produced quite close to PDD. In this case, the wind is estimated from the radar scans because they correspond to low altitude, at about 0.5 m s^{-1} . Thus, with such a low wind and a distance of a few kilometers, the NO_x eventually produced by the flashes was detected at PDD with a short delay. The strong concentration of the flashes and their good direction can explain a very large increase of NO_x at PDD.

3.2.2. 13 June 2003

On 13 June, the pressure was high at all levels over France. The very high temperatures at low levels maintained a potential instability in the air mass. During the day, several thundercells developed, especially over the mountainous areas. Thus, a convective system affected the area of PDD in the afternoon. Fig. 5 displays: (a) time series of both NO and NO_x , and of CG flash rates in the $200 \times 200 \text{ km}^2$ area, (b) the location of the CG flashes, and (c) the backward trajectory calculated with HYSPLIT at several levels and for 24 h. As indicated in Fig. 5c, the air flow was weak on this day, especially during the afternoon. So, the thunderstorms did not move very much, what is also indicated in a meteorological report for this day. A total of 1547 CG flashes occurred in the $200 \times 200 \text{ km}^2$ area during all the day. A large proportion of these CG flashes were positive, 36.5% for the whole period. This proportion was especially high during a few hours around 1200 UT, exceeding 50% since the rate for positive CG flashes was higher than that for negative ones between 1100 and 1300 UT (Fig. 5a). The average peak currents were -16.7 kA and 37.3 kA for negative and positive CG flashes, respectively. According to Fig. 5a, a large increase of NO_x was observed after 1430 UT, with a maximum of $304 \mu\text{g m}^{-3}$ at 1515 UT, and a short duration. In this case also, 50% of NO_x was NO . This case support also larger NO_x values for a large proportion of positive CG flashes. Fig. 5b displays the locations of CG flashes by distinguishing two periods, before the NO_x increase (0800–1500 UT) and after it (1500–1900 UT). The dots corresponding to the CG flashes for both periods are grey and black, respectively. Some of these CG flashes were produced at a few kilometers from PDD, in a southwesterly direction. The large NO_x increase occurred after a period of lightning activity characterized by a large proportion of positive CG flashes, in the area up-flow relative to PDD according to the general flow which was southwesterly, and in conditions of weak wind since the local measurements at PDD also indicated low values of the wind velocity. Between 1430 and 1600 UT, corresponding to the high values of NO_x , the wind velocity was lower than 2.5 m s^{-1} , while its direction changed very much in that period and was globally westerly. In this case, the production by flashes may be a cause of the very large NO_x increase.

3.2.3. 21 July 2003

The meteorological conditions of this day exhibited a strong southwesterly flow at the front of a trough of low pressure located over the close Atlantic Ocean. A convergence at low levels, because of a low of pressure over Mediterranean Sea and of a persistent altitude anomaly with warm air at the surface, maintained stormy conditions all the day,

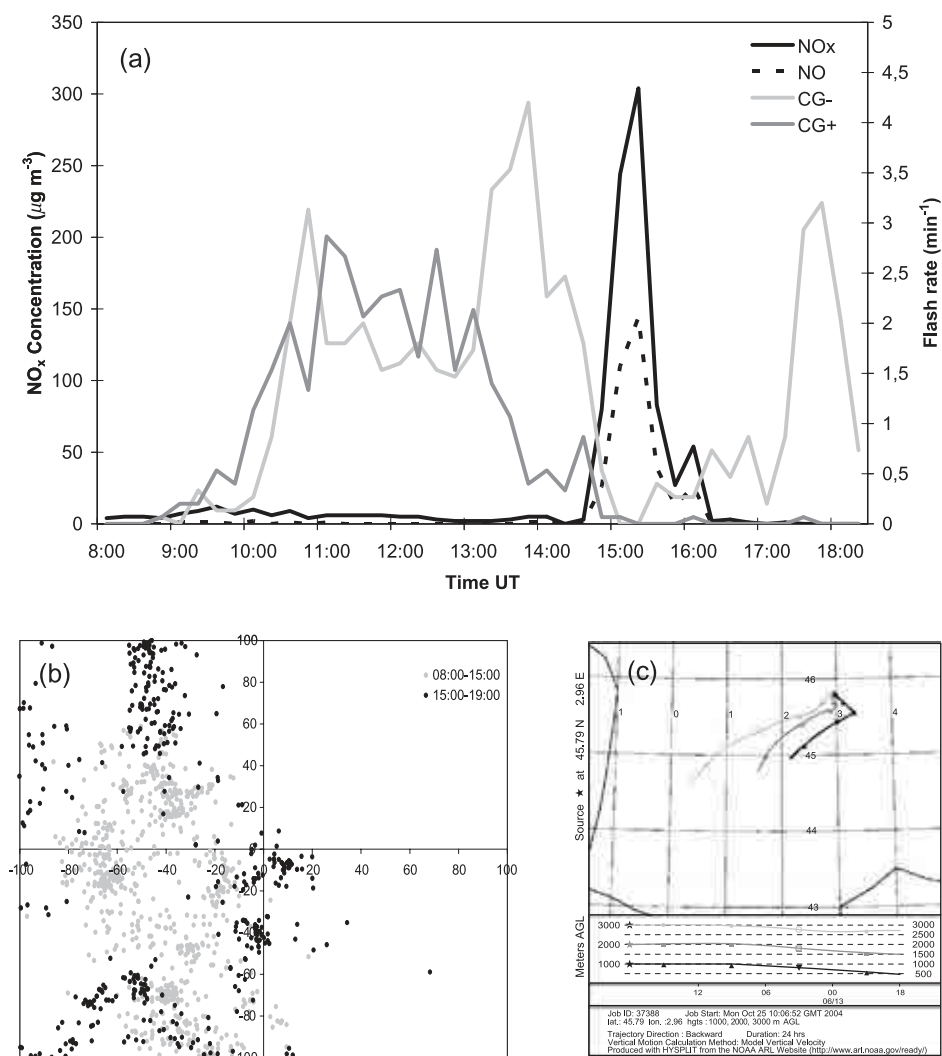


Fig. 5. Same as Fig. 3 for 13 June.

especially in Central France. So, close to the site of Puy de Dôme, several periods of lightning activity were observed. Fig. 6a displays for all the day, the time series of both NO and NO_x, and of the CG flash rates in the 200×200 km² area, by distinguishing both polarities. The NO_x clearly exhibited three rises decreasing in amplitude: 90 μg m⁻³ at 0145 UT, 66 μg m⁻³ at 1000 UT, and 62 μg m⁻³ at 1915 UT. Some values were missing because of problems of measurement. The NO proportion was high, especially for the second period with values higher than 50%. Each one of the increases was preceded by a period of lightning activity in the area. So, the flash rates exhibited several increases, around 0115 UT, between 0600 and 1000 UT and at 1545 and 1800

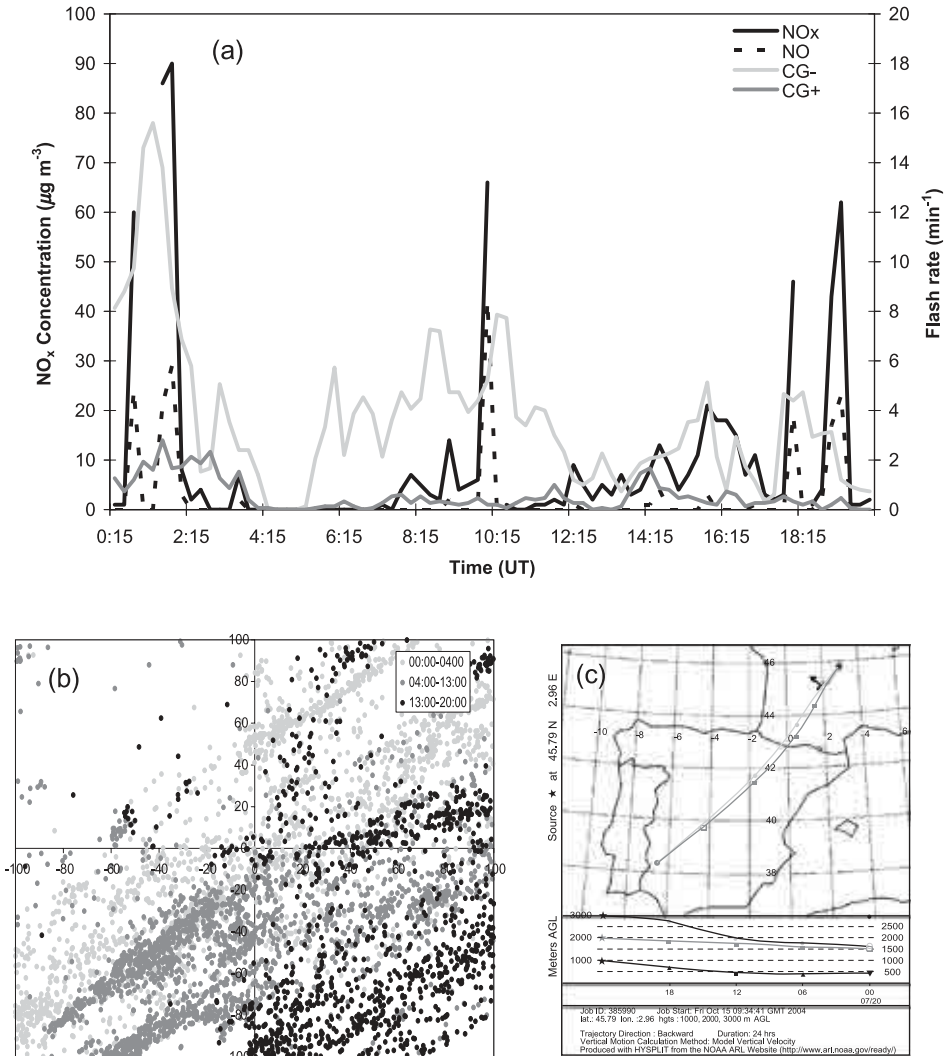


Fig. 6. Same as Fig. 3 for 21 July.

UT. The interpretation of the NO_x increases by lightning production can be supported by the observations, in terms of chronology and of amplitude. From Table 1, we can see that the fraction of positive CG flashes ranged from 5% to 11% in the three periods, and therefore the CG activity was dominated by negative CG flashes. This proportion was higher at the end of the activity for each period, as generally observed (Fuquay, 1982). Fig. 6b and c display the location of all CG flashes occurring during the day and the backward trajectory at three altitudes and for 24 h before 0000 UT on 21 July, respectively. A large amount of CG flashes were produced in the vicinity of PDD and in the southwest quarter of the 200×200 km² area. The flow was southwesterly at all

levels. All these observations can also support the production of NO_x by the flashes which might be detected at the PDD in the three periods of the day.

In order to analyze in detail the three periods of activity, Fig. 7 displays the radar reflectivity field in the same area at several instants of the day, at 0100 UT for the first period (Fig. 7a), at 0630 and 1000 UT for the second period (Fig. 7b and c), and at 1630 UT for the third period (Fig. 7d). These horizontal scans are chosen to

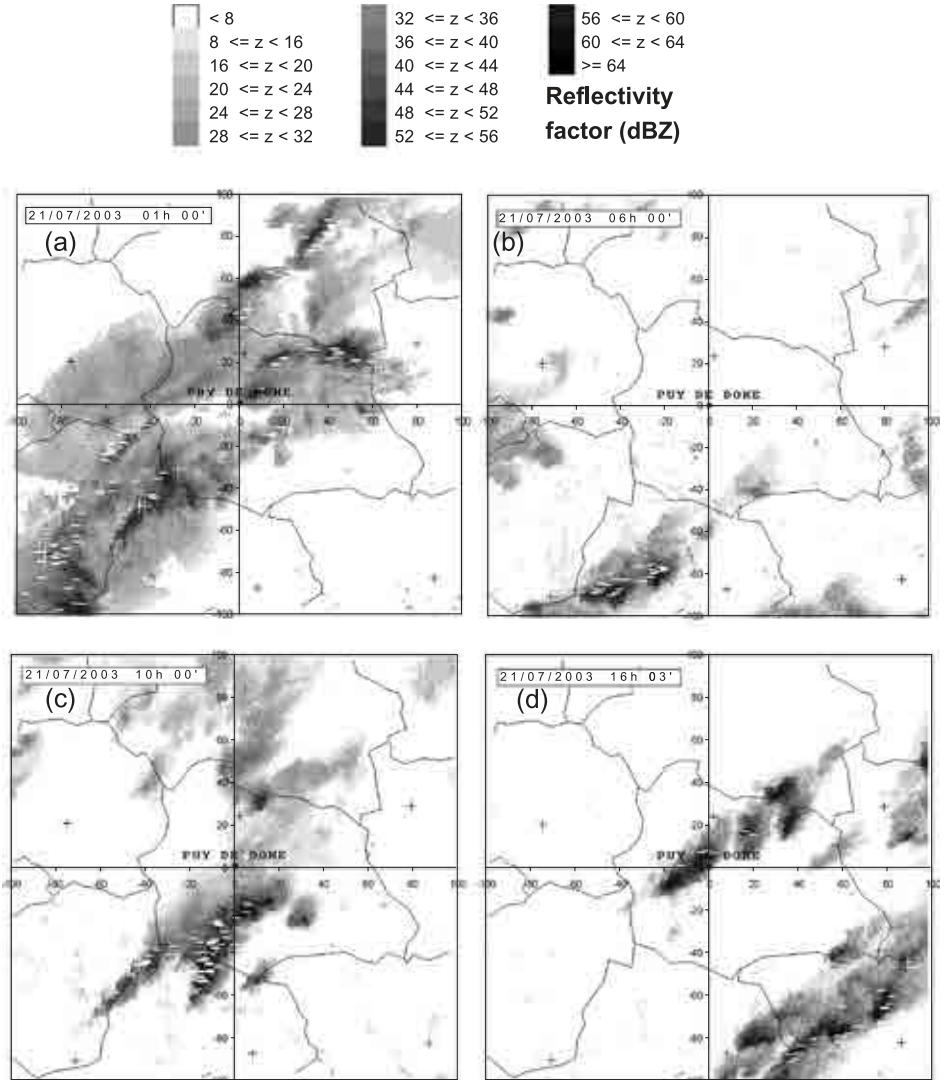


Fig. 7. Radar reflectivity horizontal field for 21 July in the $200 \times 200 \text{ km}^2$ area around Puy de Dôme: (a) at 0100 UT, (b) at 0600 UT, (c) at 1000 UT, and (d) at 1630 UT. The reflectivity factor is in dBZ. The CG flashes detected during 10 min around the time of the radar scan are superimposed with white symbols (+) and (-) for positive and negative CG, respectively.

correspond with maxima of the CG flash activity. The lightning flash locations are superimposed with $-$ and $+$ symbols for negative and positive CG flashes, respectively. So, the radar reflectivity reached about 60 dBZ at several locations at 0100 UT, between 60 and 64 dBZ at the other periods, but in these cases, the larger reflectivity values were very local. The location of the CG flashes can help to interpret the time lags between the rises of NO_x and the flash rate increases. For the first rise at 0145 UT, the CG flashes were produced almost in the southwest quarter of the area, and at a distance of several tens of kilometers of PDD but in the direction of the flow. Some flashes were also produced close to PDD corresponding to a convective system generating high reflectivity values (Fig. 7a). So, the delay between lightning activity and NO_x peak value was short. We can suppose that the NO_x corresponding to this first period were locally produced or carried by the air motion. In the second period (0600–1200 UT), the delay was larger because of a larger distance of the flash location at the beginning of activity and the rise was lower because of a lower flash rate and furthermore the flash location was not exactly in the direction of the site (Fig. 7b and c). For the third period, the delay was very weak because there was a flash activity very close to the site but the rise was low because of a very low flash rate in this case (Fig. 7d).

3.2.4. 13 August 2003

On 13 August, the pressure was high over France. The temperatures were very high and even scorching. A trough of pressure was developing north of England, organizing a weak northwesterly flow over France during the day and carrying cold air at low level on the western French coasts. During the afternoon, the thunderstorms developed over a large part of France. Fig. 8a displays the time series of CG flash rates and both NO and NO_x during the afternoon of 13 August. A large NO_x increase occurred at 1400 UT, very quickly after the first CG flashes observed in the $200 \times 200 \text{ km}^2$ area. This increase underwent two peak values, 120 and $130 \mu\text{g m}^{-3}$ at 1415 and 1500 UT, respectively. Actually, the first CG flashes were observed very close to PDD around 1330 UT as indicated in Fig. 8b. In this graph, the CG flash locations occurring before 1530 UT, that is to say before the end of the NO_x large values, are distinguished and plotted with a grey colour. The CG flash rate during this period was very low with only 1.3 min^{-1} at 1545 UT, but according to their location close to PDD, the CG flashes could be related to the NO_x increase. The flash rate was then high around 1630 UT (6.2 min^{-1}) with several cores in the area (Fig. 8b). One of these cores was located west of PDD at about 10 km and was active around 1700 UT. A new increase of NO_x was observed at 1815 UT with a peak value at $64 \mu\text{g m}^{-3}$ and therefore with a delay of about 1 h after the lightning activity. The wind velocity measured at the meteorological station at the top of Puy de Dôme was about 5 m s^{-1} in the afternoon, and according to the backward trajectories displayed in Fig. 8c, the air motion was northwest–westerly and therefore could transport the NO_x produced in this lightning core. For this event, the proportion of positive CG flashes was rather high, 36% for the whole period. It was especially high during the first NO_x increase between 1400 and 1530 UT, corresponding to the higher NO_x values, since the positive CG flash rate exceeded that of the negative CG flash at 1415 and after 1500 UT.

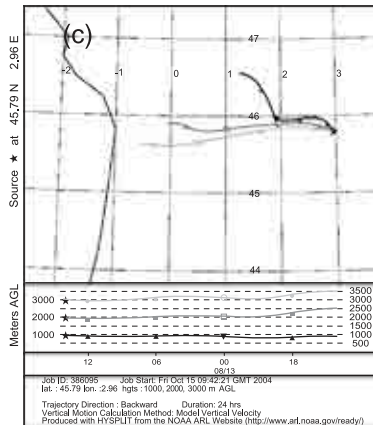
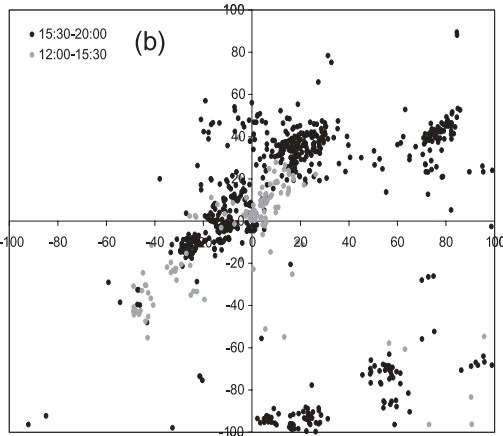
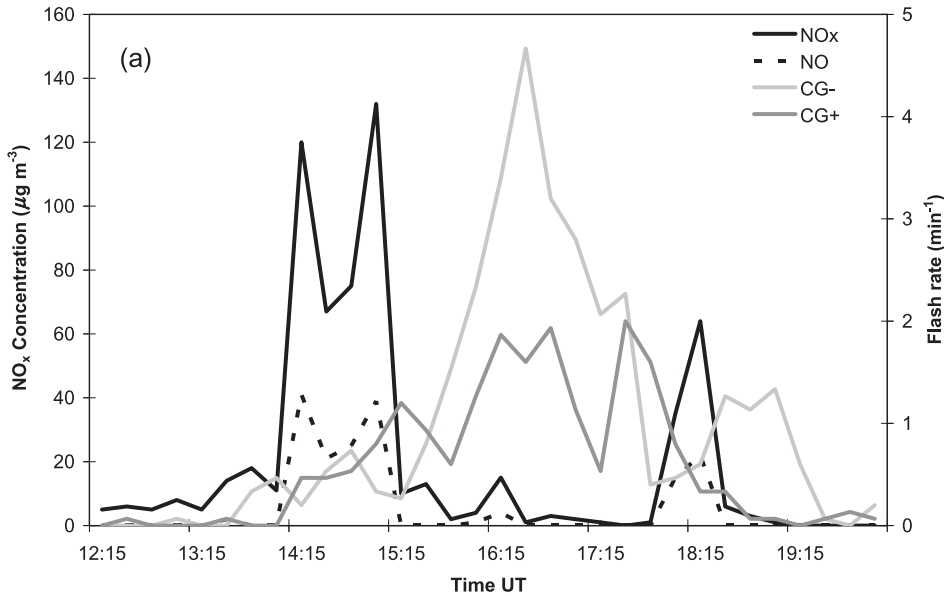


Fig. 8. Same as Fig. 3 for 13 August.

3.2.5. 28 August 2003

A deep trough of low pressure was located over the nearby Atlantic at the northwest of the Iberian Peninsula on 28 August. A strong southwesterly flow was established over France and made unstable warm air masses at low levels. Two altitude anomalies succeeded during the day and a strong thunderstorm activity crossed the whole country. In the $200 \times 200 \text{ km}^2$ area around PDD, 8045 CG flashes were recorded by Météorage during all the day. However, according to Fig. 9b, most CG flashes were produced far from PDD. The global proportion of positive CG flashes was 10% for this day, which is a typical value for the French latitudes (Soula and Chauzy, 2001). The positive CG rate remained

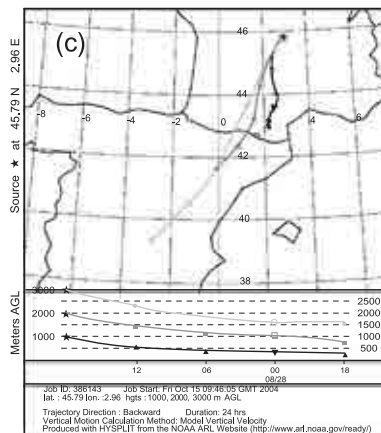
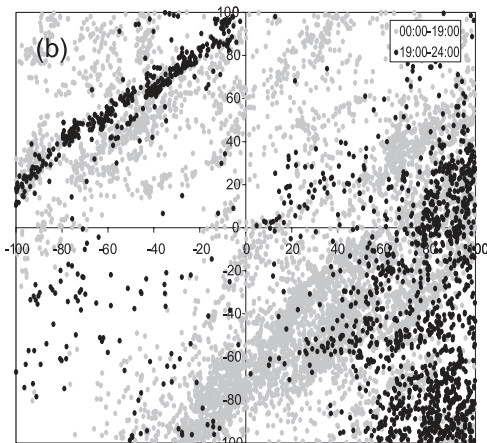
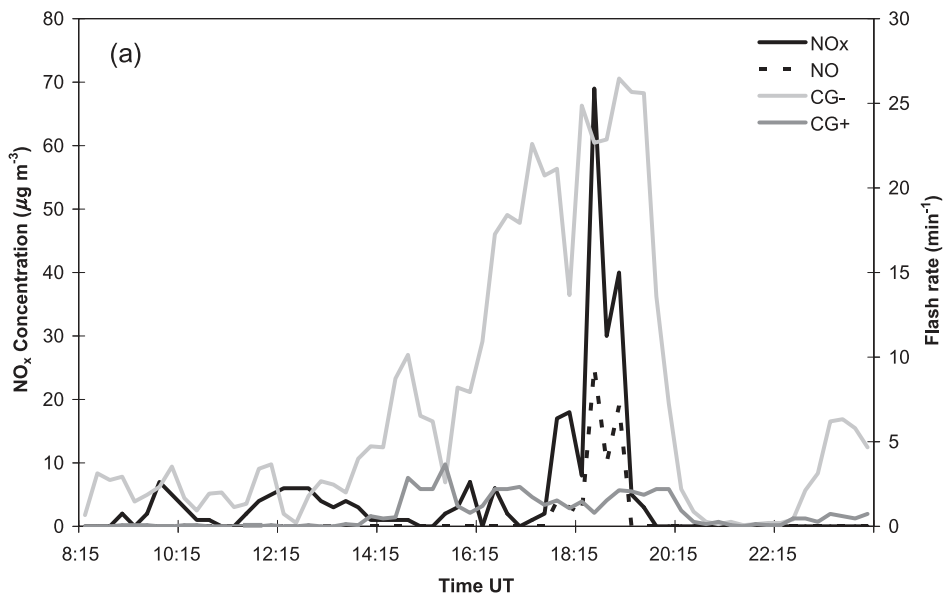


Fig. 9. Same as Fig. 3 for 28 August.

much lower during the whole activity, especially during the period of high NO_x values. According to Fig. 9a, a rather large increase of NO_x was observed around 1830 UT with a maximum value of $69 \mu\text{g m}^{-3}$, but some small increases occurred before. The proportion of NO during the large NO_x increase was high, with about 40%, contrary to the small NO_x increases. It confirms the presence of a rather local source. The flash rates and the NO_x seemed well correlated. The location of the CG flashes which occurred before the end of the main NO_x increase, that is to say before 1900 UT, are plotted with grey dots in Fig. 9b. These CG flashes were potentially able to affect the NO_x at PDD in inssofar as they are well

located according to the air flow. The backward trajectories displayed in Fig. 9c indicate a south–southwesterly averaged flow at several levels. The local wind measurement at PDD provided a direction fluctuating between a southerly and westerly direction during the main NO_x increase and values from about 2 m s^{-1} at 1800 UT to 10 m s^{-1} at 2000 UT. All this information about the air flow shows that a large number of CG flashes were really able to affect the NO_x at PDD.

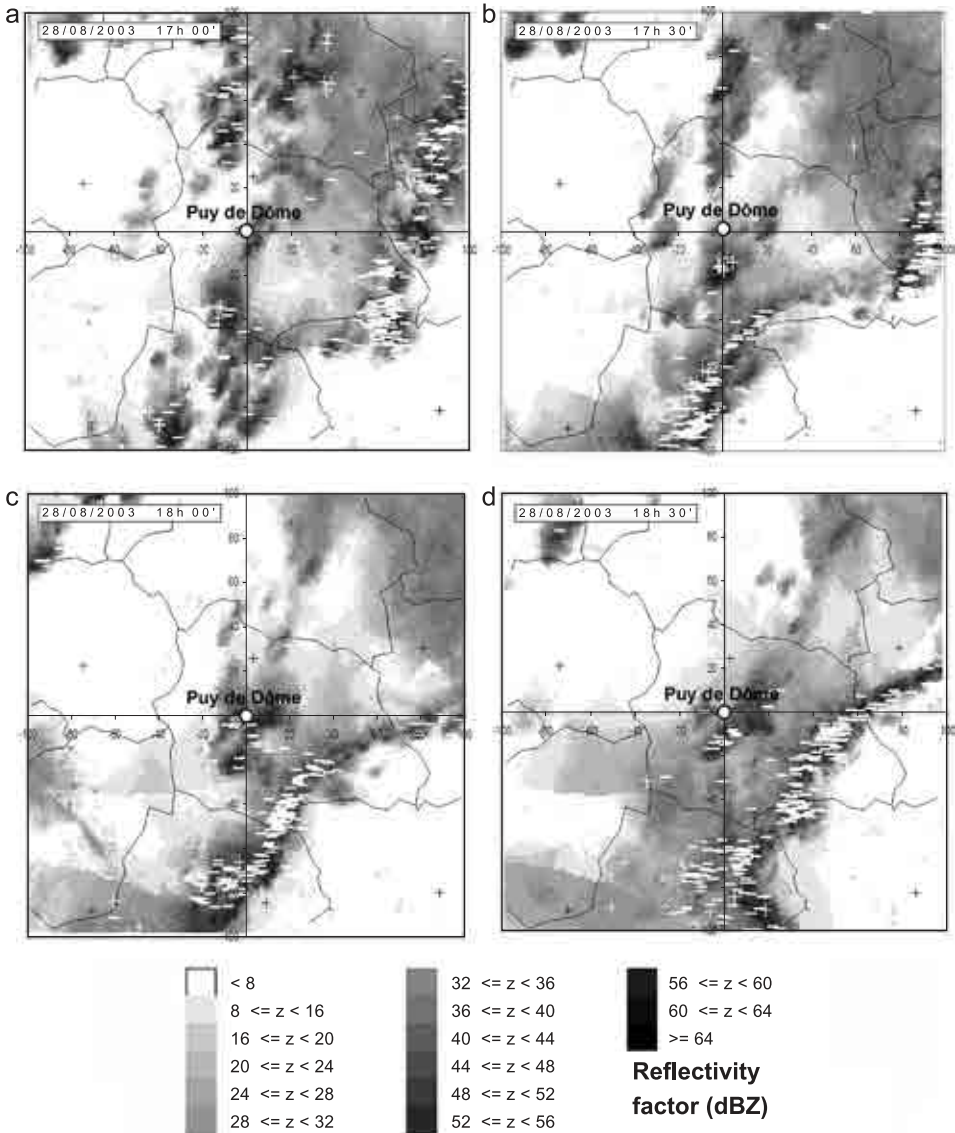


Fig. 10. Same as Fig. 7 for 28 August. The times are (a) 1700 UT, (b) 1730 UT, (c) 1800 UT, and (d) 1830 UT.

Since the CG flash activity was very strong on this day, up to 28.6 min^{-1} for the whole area at 1900 UT, it is interesting to analyze in detail the flash location during the main NO_x increase. Fig. 9b distinguishes the CG flashes produced before 1900 UT, that is to say the end of the large NO_x values but as their number was really large, Fig. 10 displays over shorter periods their location superimposed on the radar images. It corresponds to 4 images at 1700, 1730, 1800, and 1800 UT, with for each one, the CG flashes over 10-min periods. The system displacement velocity and direction can be deduced from these graphs and

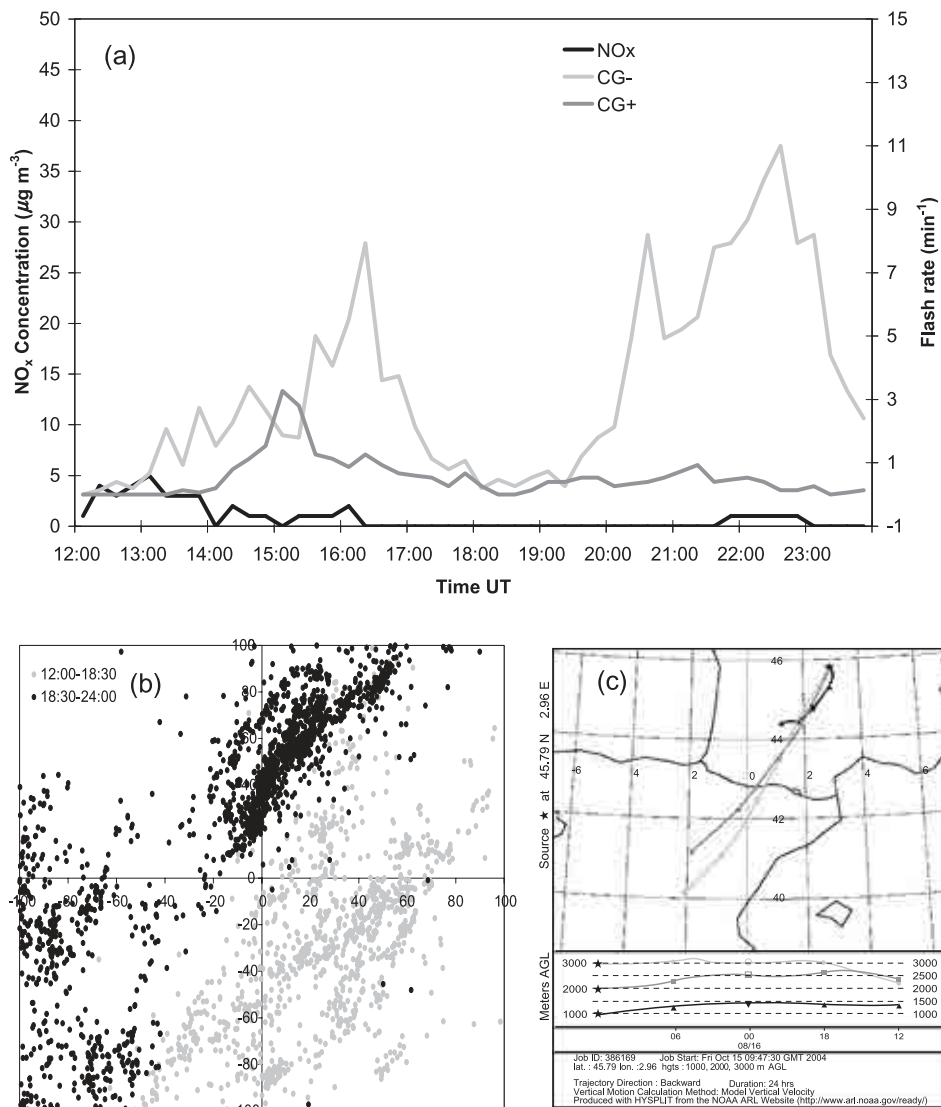


Fig. 11. Same as Fig. 3 for 16 August.

they are estimated to be 12 m s^{-1} and south to southwesterly, respectively. Even if most of the flashes did not constitute a possible source of NO_x for PDD because they were located too far to the southeast, the closer CG flashes were produced around 1800 UT, that is to say when the NO_x started to significantly increase. This complement of information about the convective system confirms the possible NO_x source by the CG flashes.

3.2.6. 16 August 2003

The meteorological situation of this day was characterized by the presence of a trough of pressure over Atlantic and Northern Spain with a southwesterly flow with anticyclone curve at the front. A first convective system went up Spain and went on over France during the night. The thunderstorm activity started again in the afternoon to become strong in the evening over several areas of South-Western, Central, and Eastern France. The $200 \times 200 \text{ km}^2$ area was struck by 2849 CG flashes during all the day, but without any flash really close to PDD (Fig. 11b). The most active system was north of PDD and active after 1830 UT and the closer flashes occurred between 1600 and 1700 UT at about 10 km south of PDD and not of high density. The proportion of positive CG flashes was 14%, therefore close to the typical value. Fig. 11a displays the time series of NO_x and CG flash rates. NO_x did not reach large values and did not undergo any increase associated with lightning activity. During this day, the maximum value was $5 \mu\text{g m}^{-3}$ at 1300 UT. The flash rate evolution indicated two active periods, one during the afternoon and the other in the evening. The total CG rate reached its maximum values at 1615 and 2230 UT, 9.2 and 11.1 min^{-1} , respectively. Since the flow was really southwesterly, which is confirmed with the backward trajectories (Fig. 11c) and with the local wind measurements, it seems logical that any increase of NO_x was visible in the time series. Actually, when the closer CG flashes occurred after 1600 UT, the local wind velocity increased close to 10 m s^{-1} and the direction turned from easterly to southwesterly.

4. Discussion and conclusion

Large NO_x increases observed at a station at high altitude have been analyzed in relation with the lightning activity provided by the CG location network Météorage, the characteristics of the air flow provided by a trajectory calculation model and local wind measurements, and for some cases with radar images provided by the French ARAMIS network. The analysis period is 4 months in 2003, from May to August. During this 4-month period, 16 days and 7 days are affected by NO_x peak values exceeding $50 \mu\text{g m}^{-3}$ and $100 \mu\text{g m}^{-3}$, respectively. Of the 16 days, 14 (87.5%) correspond to lightning activity in the $200 \times 200 \text{ km}^2$ area around PDD and of the 7 days, all correspond to lightning activity in this area. On the other hand, 33 days of the period are affected by lightning activity in the PDD area of which only 14 (42%) correspond to NO_x peak values exceeding $50 \mu\text{g m}^{-3}$. The presence of lightning activity seems to be a necessary condition to have very large peak values of NO_x at PDD, but this condition was not sufficient to ensure high values. According to the case studies analyzed in Section 3.2, large NO_x values were observed at PDD when the CG lightning flashes occurred either very close to the site, or upstream of the station. Another parameter seems interesting to point out, that is the positive CG flash proportion.

From Table 1, summarizing the main characteristics of each case, it is clear that the higher NO_x values are associated with the higher positive CG flash proportion and simultaneously with lower wind conditions. It seems consistent that the high NO_x values can be associated with low winds because of lower dispersion. A relation with the proportion of positive CG flashes could be due to the higher associated peak currents. Since the LNO_x production is associated with the energy dissipated in the lightning channel, high currents of positive CG return strokes could be more efficient in this mechanism of production.

The time delay between the NO_x increase and the CG flash occurrence is relatively consistent with the CG location and the characteristics of the air motion observed. The case of 21 July with three periods of lightning activity in the area (Section 3.2.3) illustrates very well this phenomenon. The first and third periods of this case correspond to CG flashes close to PDD with short time delays between NO_x detection and the flash, while the second period corresponds to CG flashes far away and upwind from PDD with a large time delay. The case of the highest NO_x peak value ($330 \mu\text{g m}^{-3}$) is 31 May corresponding to a very low CG lightning activity. The isolated thunderstorm which produced the lightning flashes was very close to PDD and the wind was low at that moment. The radar images show that the system was quasi-stationary and consequently, the local CG flash density was not locally low. A large proportion of the CG flashes were positive. In this case, the local weak wind conditions, the proximity and the rather high density of the CG flashes could explain the very large NO_x values. Other cases are analyzed in detail for the correspondence between the NO_x increase and the CG flash activity, and they confirm the possible explanation by lightning production. So, the case of 13 August accounts for a first increase of NO_x rapidly after the first CG flashes produced very close to the station and a second one with a delay due to remote CG flashes located west of PDD. The case of 28 August supports both the transport from remote CG flashes and the local production by some flashes. The case of 16 August, during which a lot of CG flashes occurred in the area, shows that if the CG flashes are not produced close to PDD or in the good direction related to the wind, any large NO_x increase is not observed.

For all cases studied in detail, the air flow did not correspond to meteorological conditions carrying polluted air from the big city of Clermont–Ferrand located 10 km east of PDD. Most of the time, it was southwesterly and sometimes westerly; however, for some cases, the local wind was fluctuating or was not available. So, in order to estimate the ability of the transport of a plume from the Clermont–Ferrand metropolitan area located east of PDD, the values of NO_x measured at other stations in the area have been analyzed in the cases of large peak values at PDD. These data are available on the site (www.atmonet.org). For example, on 31 May in the city area, at several urban or suburban stations, the hourly average of NO_x did not exceed $40 \mu\text{g m}^{-3}$. This value is far from those observed at PDD, about $200 \mu\text{g m}^{-3}$ for the hourly average. Furthermore, at these stations, NO_2 was largely dominant while at PDD, NO and NO_2 were close during the large increase of that day. The second example, 13 June, illustrates also this difference between the large values observed at PDD and those observed in the stations scattered in the city area. On 13 June, the maximum NO_x hourly average observed at an urban station was about $60 \mu\text{g m}^{-3}$ with a large proportion of NO_2 and it was more than $120 \mu\text{g m}^{-3}$ in an hourly average at PDD with close proportions of NO and NO_2 . In each case study

considered, the NO proportion was high during the large NO_x increases observed at PDD, which indicates a probable local source. The NO_x constituents are therefore very different in PDD measurements and in the urban area. Pollution by this city cannot explain the presence of large NO_x concentrations at PDD. The CG lightning flashes produced by thunderstorms in the area around PDD seem therefore to produce substantial amounts of NO_x which could significantly affect the measurement at PDD.

The highest value obtained during the summer of 2003 was 330 μg m⁻³ and it is well associated with a thunderstorm stationary and close to the measurement site. Furthermore, this value was not isolated since it was preceded by several other high values close to 300 μg m⁻³. No information about the IC flash activity was available and therefore it is difficult to know if the NO_x measured were of high or low altitude in origin. Such values around 300 μg m⁻³ correspond to values between 176 ppbv and 270 ppbv according to the respective proportion of NO₂ and NO. The NO_x values observed in stormy conditions at the ground can be therefore comparable to those measured at altitude by aircraft and even much larger at some moments (Höller et al., 1999; Stith et al., 1999). It can indicate that the return stroke of the CG flash is an important phase for the production of NO_x. The fact that the positive CG flashes seem to produce more NO_x is also consistent with this process because the peak current is generally higher for positive flashes. The cases of 31 May and 13 June providing these high NO_x values around 300 μg m⁻³ also exhibit high peak current values since the mean values were 42.9 and 37.3 kA for positive CG flashes. These cases give a ground observation proof of LNO_x. It would be necessary to use several measurement stations and numerical models for a quantitative determination of the amount of NO_x produced by a given length of lightning channel.

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