Nonequilibrium calculations of the role of electron impact in the production of NO and its emissions

Laurence Campbell and Michael J. Brunger
ARC Centre for Antimatter-Matter Studies, School of Chemistry, Physics and Earth Sciences, Flinders University, GPO Box 2100, Adelaide SA 5001, Australia
E-mail: laurence.campbell@flinders.edu.au

Abstract. We review our recent work on nonequilibrium modelling of the density of nitric oxide and its infrared emissions in the Earth’s upper atmosphere. The aim of these studies was to investigate the contribution of electron impact excitation to the NO density and the sensitivity of this process to the electron impact cross sections. The results are compared with satellite measurements of NO densities in equatorial and auroral high-latitude conditions and with rocket measurements of infrared emissions in auroral conditions. Particular findings are that electron impact excitation of $\text{N}_2$ makes a significant contribution to the NO density at altitudes around 105 km and to auroral infrared emissions for the $\text{(1} \rightarrow \text{0)}$ ground-state emission from NO. The sensitivity of the NO fundamental emissions to various measured and theoretical integral cross sections is investigated and found to be significant.

1. Introduction
Electron impact (photoelectron and auroral) in the Earth’s upper atmosphere plays a major part in the production of nitric oxide [1] and in infrared emissions from this species [2]. The main production mechanism is electron impact ionisation and dissociation of atmospheric species, which produces $\text{N(}^2\text{D)}$ (excited N) that then reacts with $\text{O}_2$ to produce NO. A minor source is electron impact excitation of $\text{N}_2[A^3\Sigma_u^+]$, which reacts with $\text{O}$ atoms to produce NO and $\text{N(}^2\text{D)}$ [3]. This $\text{N(}^2\text{D)}$ reacts with further $\text{O}_2$ to produce even more NO, enhancing the effect of this excitation source. The NO produced by $\text{N(}^2\text{D)} + \text{O}_2$ is vibrationally excited, leading to chemiluminescent infrared emissions. Recently direct electron impact excitation has also been shown (in calculations) to contribute to infrared emission by NO [4].

In order to investigate the importance of the $\text{N}_2[A^3\Sigma_u^+]$ source of NO, a model which predicts NO density in the atmosphere was developed [5, 6]. Above 130 km the NO density reaches a chemical equilibrium value that varies with solar insolation and auroral input. NO is transported downwards by molecular diffusion at altitudes above about 110 km and by eddy diffusion below this height. Vertical winds produced by auroral heating may also redistribute NO vertically. As a result of these transport processes the NO density does not reach a chemical equilibrium value at altitudes below 130 km, so a nonequilibrium calculation is required to predict the NO density in the region of its peak at around 110 km.

The model has been applied to predict the NO density profile in the upper atmosphere at the equator, and at 65° N with moderate auroral input [6]. It was run with and without the $\text{N}_2[A^3\Sigma_u^+] + \text{O}$ reaction and in both the equatorial and high-latitude cases it was found that
the contribution of this reaction is significant, and that the predicted densities are closer to the observed values when the N₂[A³Σ⁺⁺] contribution is included.

The model was then used to calculate the background NO density as part of an investigation of infrared emissions from NO [7]. Auroral electron impact produces such emissions principally by chemiluminescent emission from NO* (vibrationally excited NO) that is produced by the N + O₂ reaction [8]. However, measurements [9, 10] of electron impact excitation of the first three vibrational levels of NO made possible calculations of the contribution of electron impact to the infrared emissions. It was found that the contribution is significant relative to chemiluminescence for the ν’ = 1 level, while the smaller contribution for the other levels may nevertheless alter the spectrum from that of chemiluminescence in a way that could be exploited in remote sensing [4]. The nonequilibrium model to calculate NO density allowed a detailed calculation [7] of these electron-driven infrared emissions for the particular conditions of a rocket measurement [8]. It allowed both a calculation of the background NO density for the time and place, and of the increase in the density due to the auroral input. While there was a discrepancy between the absolute value of the predicted emissions and the experiment, it was found that the shape of the predicted spectrum of NO fundamental emissions matched the measurements better when direct electron excitation was included. This simulation of the rocket measurements was recently repeated [11] with new measurements [12] and theoretical calculations [13] of electron impact vibrational excitation of NO. In both cases the agreement with the measurements is improved for the ν’ = 1 level, particularly using the new theoretical cross sections. However, the new values lead to an overestimation for ν’ = 2.

2. Computational methods

Two computational methods are used in this work. A time-step simulation, in which the change in density of each species is calculated for a small time step, is used for slower reactions. For species which reach equilibrium quickly, a statistical equilibrium calculation is used.

In the time-step calculation the incremental change in density ∆[z] for each species z is calculated at successive small time steps of ∆t as ∆[z] = R[xy][z]Δt, where R is the rate for the reaction x + y → z. To implement this computationally, all density changes in the time step for each species z are summed, so that positive and negative increments cancel before the densities are adjusted for all species at the end of the time step.

Alternatively, for fast reactions, the positive and negative contributions to ∆[z], ∆[zp] and ∆[zn], are summed separately. The next value of [z] is calculated by solving the continuity equation, in which the gain ∆[zp] is assumed equal to the new value of the density [z] multiplied by the loss rate in the current interval of ∆[zn]/[z], i.e.

\[ [z]' = \frac{\Delta[z_p][z]}{\Delta[z_n]} \]  

Equation (1) is applied for all species being considered, and iteration of this process continues until the populations of all species reach “statistical equilibrium” [14].

In some cases, such as the determination of the population densities of the excited states of N₂ in the atmosphere, produced by photoelectrons and auroral electrons, there is a wide range of excitation probabilities and radiative transition probabilities. This large range, together with the large number of different pathways, makes a time-step calculation impractical and the statistical equilibrium calculation is used. Equation (1) is applied for all electronic states being considered, and iteration of this process continues until the populations of all states reach equilibrium. The population densities of the excited states of N₂ are then imported into the time-step calculation.

There is a wide range of different rates of chemical reactions in the upper atmosphere. While a time-step calculation at 1-s intervals is suitable for most reactions, those that are too fast
for this interval must be identified and the equilibrium calculation performed. The issue of combining the two approaches has been described previously [15].

3. Atmospheric model
The current atmospheric model has its origin in a study of the role of excitation of the $N_2[A^3\Sigma^+_u]$ state in setting the electron density in the upper atmosphere [5]. Inputs required by the original model from other work were:

- densities of $N_2$, $O_2$, and $O$ atoms from the MSISE90 [16] model,
- an established model of photoionisation and photodissociation by sunlight [17], and
- measured NO densities [18, 19].

A flux distribution for auroral electrons was developed [5] by combining various measurements and model results.

The photoionisation and photodissociation produced by sunlight, plus ionisation and dissociation produced by photoelectrons and auroral electrons, are calculated for particular solar and auroral conditions. The ions and electrons produced take part in a series of other reactions, as outlined by Barth [1]. These processes were modelled in a time-step calculation which allows for nonequilibrium conditions, such as solar input varying with time of day and short-duration aurora. Using a time-step simulation also allows the inclusion of molecular diffusion downwards at higher altitudes and eddy diffusion, also downwards, at lower altitudes.

Simultaneous calculations of the populations of the excited state and ground state vibrational levels of $N_2$ allowed the importance of the role of the $N_2[A^3\Sigma^+_u]$ state to be calculated. It was found that the contribution of the $N_2[A^3\Sigma^+_u]$ state to the population levels of the ground state was not as important as the higher electron temperature produced by auroral heating. Nevertheless the model produced a range of values for atmospheric parameters that may be compared with other models or measurements.

The model was then used to calculate profiles of electron density in the atmosphere, using two different sets of reaction rates for the reaction

$$O^+(4S) + N_2(\nu'') \rightarrow NO^+ + N(4S)$$

which is critical in setting the electron density due to the high recombination rate of NO$^+$ with electrons. The predictions in each case were compared with those of the International Reference Ionosphere (IRI) [16]. It was shown [20] that the $\nu'' = 0$ rate of St. Maurice and Torr [21], enhanced for $\nu'' > 0$ by factors determined by Schmeltekopf et al. [22], led to predictions of electron densities in better agreement with the IRI than predicted using the reaction rates of Van Zandt and O’Malley [23].

The model was then modified [15] to calculate the NO densities in the upper atmosphere. It was applied to the specific nonequilibrium case of a 4.5-day simulation of sunlight plus moderate auroral activity, with 1 h of strong auroral activity at the end. This simulation predicted a value of the NO density at 106 km that was consistent with the maximum density measured by the SNOE (Student Nitric Oxide Explorer) satellite [24]. While this consistency was not definitive, as the duration of the strong aurora was arbitrarily chosen, it justified further development of the model for the recent work reviewed in the next section.

4. Recent Applications
The model described above was applied to predict atmospheric NO densities [6] in situations where averages of satellite measurements were available. As part of this study it was necessary to include a new reaction rate and consider the effect of vertical winds.

It was then applied to make predictions of NO infrared emissions [7] for the specific conditions of a rocket measurement. This required both the nonequilibrium calculation described above to
predict the background NO density for the location, then another nonequilibrium calculation of
the increase in NO due to the extra auroral activity at the time of the rocket flight.

Finally the predictions of the rocket measurements were repeated [11] for recent measurements
and recent theoretical calculations of the integral cross sections for electron impact vibrational
excitation of NO, to investigate the sensitivity of the emissions to the cross sections.

4.1. Nonequilibrium calculations of NO density

Average values of NO density in the upper atmosphere at 11:00 LT have been determined [24]
from measurements by the SNOE satellite for several intervals around the spring and autumn
equinoxes, for equatorial conditions and for moderate geomagnetic conditions at 65° N
geomagnetic latitude.

The model described previously [15] was used, but updated with some recent reaction rates
for,

\[ \text{N}(^2\text{D}) + \text{O}_2 \rightarrow \text{NO} + \text{O} \quad \text{(R2)} \]

\[ \text{N}(^4\text{S}) + \text{O}_2 \rightarrow \text{NO} + \text{O} \quad \text{(R3)} \]

As reported previously [15] molecular and eddy diffusion of NO and N(^4S) were included.
Details of the parameters used for these transport processes were detailed later [6], including
the extension to the equatorial case.

As the atmospheric model was based on one by Barth, a comparison is made in figure 1 of the
current calculations with previous predictions by Barth [1]. The quantity compared is the rate
of NO production by the reactions N(^4S) + O_2 and N(^2D) + O_2. There is substantial similarity,
giving credibility to the current model.

Also plotted in figure 1 are the production rates of NO due to the reactions

\[ \text{N}_2[\text{A}^3\Sigma_u^+] + \text{O} \rightarrow \text{NO} + \text{N}(^2\text{D}) \quad \text{(R4)} \]

\[ \text{N}_2[\text{X}^1\Sigma_g^+](\nu' > 11) + \text{O} \rightarrow \text{NO} + \text{N} \quad \text{(R5)} \]

While these reactions contribute less than one tenth of the N-atom contribution, reaction (R3)
is nevertheless significant because it produces excited N atoms, which can then react with O_2 to
produce even more NO. The model was run with and without the excited N_2 to quantitatively
investigate the effect of reactions (R4) and (R5).

The nonequilibrium nature of the model is illustrated in figure 2. It is run for 101 hours,
starting at 6:00. The calculated NO densities are shown (horizontal axis) as a function of altitude
at 11:00 on each successive day. While equilibrium is reached quickly above 150 km, it takes
two days for the NO density to build up to a near constant value at around 110 km. This is
a non-physical example because the NO density does not start at zero. However, it illustrates
the time constant of the processes acting. (An early estimate of NO densities by Mitra [27] is
included to illustrate the deficiency of an equilibrium calculation for NO density).

Figure 3 shows the relative importance of the contributions of excited N_2 and the enhanced
rate of reaction (R2), which is greater than in the earlier modelling. While the full model gives
densities in fair agreement with the measurements (except around 120 km), the calculated NO
densities get smaller as the model is run progressively without the contributions of N_2[A^3Sigma_u^+],
N_2[X^1Sigma_g^+](\nu' > 11) and the enhancement of the rate for N(^2D) + O_2. The contribution of
vibrationally excited N_2 is small, particularly at the lower altitudes, while the contribution of
N_2[A^3Sigma_u^+] is more significant than the enhancement of (R2) in the region of the peak NO density.
The enhancement of the N(^2D) rate is relatively more significant at higher altitudes.

Similar results are shown in figure 4 for the auroral region. SNOE measurements are
shown for high geomagnetic activity at 65° N geomagnetic latitude and for the highest density
measured. The simulation in this case is run for 77 hours, which is a reasonable duration for high
geomagnetic activity, starting at 6:00. The model is run for auroral fluxes of 0.5 erg cm^{-2}s^{-1} and
1.0 erg cm^{-2}s^{-1}, the latter being the value chosen by Barth [1]. The calculated NO densities are
close to the average values for the lower auroral flux, while agreement with the highest density at 106 km is given by using the higher auroral flux. As for the equatorial case, the calculated NO densities are substantially less (by a factor of ~3) in the region of peak density when the $N_2[A^3Σ_u^+]$ contribution is excluded.

In figure 4 a simulation of Barth’s predictions [1] was made by running the current model (without the excited $N_2$ contribution and the two updated reaction rates) for the conditions of Barth’s calculations. The predicted NO densities agree well with those of Barth, giving credibility to the current model and the main conclusion that excited $N_2$ must be included in order to produce reasonable agreement with the measurements.

As part of this study the role of vertical winds was investigated, as such winds can be produced by auroral heating. To investigate whether vertical winds can affect the calculated NO densities, a measured wind record [28] at 120 km for auroral conditions was applied in the simulation. This record (shown in figure 5) is for one night, so it is repeated on successive nights and the daytime values are interpolated as shown in figure 5. This gives a predominately downward wind prior to the SNOE measurement time of 11:00. As the wind measurements are only for one height, the vertical wind values at heights above 120 km were found by scaling [7] based on other observations in which vertical winds in auroral conditions were measured at two heights simultaneously [29]. Below 120 km, assuming conservation of mass, the speed is scaled as the reciprocal of the atmospheric density. The speeds were divided by 20, given that the

Figure 1. Comparison of current calculations (●) with results of Barth [1] (——–) for production rates of NO due to reactions involving species as labelled.

Figure 2. A comparison of the calculated NO density at 11 am on successive days of the simulation with the SNOE measurements for high (——–) and moderate (——–) solar activity and with an empirical model (◊). Error bars at 122.5 km show the estimated measurement uncertainty and the altitude range for the high solar activity case. The single horizontal error bar shows the estimated uncertainty due to solar input at 122.5 km to the calculated density for 107 hours.
auroral flux of 0.5 erg cm$^{-2}$s$^{-1}$ that we assume is likely to be much less than that at the time of the wind measurements. Wind is included in the model by calculating, in each time step, the change in density in each height interval due to the amount of NO that is added or removed at the top and bottom of the height interval by the wind.

In figure 6 the calculated NO densities are shown for the application of the “downward” vertical wind just described, and for an “upward” wind case implemented by taking the inverse of all the wind speeds. As expected, the downward wind transfers NO from higher to lower levels while the upward wind transfers it the other way. However, the effect is not so large as to change the overall shape of the density profile with height, and in practice it is to be expected to be smaller due to averaging of the effects of upward and downward vertical winds over the time and space of the SNOE measurements. The addition of the effect of vertical winds illustrates how the nonequilibrium nature of the current model allows the effect of other parameters to be easily included and studied.

4.2. Nonequilibrium calculations of NO infrared emissions

Nonequilibrium calculations are required to predict NO infrared emissions during auroral conditions [15]. The NO density increases with time due to NO production by auroral electron impact. Also the temperature and density of the thermal electrons in the atmosphere increase with auroral input, so the electron-impact induced vibrational excitation of the NO increases [15].

The model described above has been modified to enable prediction of infrared emissions from
Figure 5. Measurements of vertical neutral wind (positive upward) [28] with low (— — —) and high (· · ·) error and the fit (———) to these measurements used in this work. The time of the NO measurements (11:00 LT) is indicated.

Figure 6. A comparison of calculated NO densities with measurements (———) for the cases of no vertical wind (●), upward wind before the measurement time (○) and downward wind before the measurement time (△).

NO. The previously accepted sources [2] for this emission are nightglow, produced by radiative decay following the collisional excitation:

$$\text{NO}(\nu'' = 0) + \text{O} \rightarrow \text{NO}(\nu' = 1) + \text{O}$$  \hspace{1cm} (R6)

and the chemiluminescent reactions,

$$\text{N}^2(\text{D}) + \text{O}_2 \rightarrow \text{NO}(\nu') + \text{O} \hspace{0.5cm} (\nu' = 1 - 12) \hspace{1cm} (R2')$$

$$\text{N}^2(\text{S}) + \text{O}_2 \rightarrow \text{NO}(\nu') + \text{O}. \hspace{0.5cm} (\nu' = 1 - 9) \hspace{1cm} (R3')$$

Recent measurements [22] of the vibrational excitation cross sections for NO at low impact energy, i.e.

$$\text{NO}(\nu'' = 0) + \text{e}^- \rightarrow \text{NO}(\nu') + \text{e}^- \hspace{0.5cm} (\nu' = 1, 2, 3) \hspace{1cm} (R7)$$

have enabled calculations of the infrared emission due to electron impact to be made. This was first investigated with an equilibrium auroral model [4]. The development of the nonequilibrium atmospheric model has allowed simulations to be performed for specific conditions. In particular, it allowed the calculation of both the background NO density and the increase due to auroral input [7] for the conditions of a particular rocket measurement [8].

Figure 7 shows the calculated NO densities after a 71-h simulation for the time and place of the rocket measurement, again including the calculation with and without the excited N$_2$ contributions and with the smaller reaction rate for (R2). For reference the SNOE measurements are included.

The model was then run for another 3 minutes with much stronger auroral activity added and the infrared emissions calculated. Due to the speed of the processes involved (radiative decay and collisional quenching of the vibrationally excited NO) a time step of 0.001 s was required. An aurora of strength 5 kR (kRayleighs [30]) was applied, as this was the quoted strength (from ground-based observations of the 557.7 nm line) at the time of the rocket measurements. The calculated fundamental emissions, i.e. (1 → 0), (2 → 1) . . . , are compared in figure 8 with the
values of these emissions deduced [8] from the measurements. The $(1 \to 0)$ emission was divided into nightglow and auroral components, with the much larger one being the nightglow.

The calculated brightness of the nightglow, chemiluminescent emissions and emissions due to electron impact are shown for the current model. There is reasonable agreement for the nightglow value around 4500 kR, indicating that the calculated NO density for the conditions of the measurements is valid. There is a large discrepancy between calculations and the measurements of the auroral emissions. However, the rocket was launched when the aurora had a strength of 50 kR, and when a value of 40 kR is used in the current calculation, it produces agreement within error for all except the $(1 \to 0)$ emission. So it may be that the quoted reduction of the auroral strength from 50 to 5 kR during the time of the rocket measurements is not appropriate to the region in which the infrared emissions occurred.

The electron impact contribution to the emissions is much smaller than that of the chemiluminescent reactions, except for the $(1 \to 0)$ emission. The addition of electron impact improves the agreement of the calculated $(1 \to 0)$ emission with the measured value, and improves the overall agreement for the shape of the spectrum.

4.3. Sensitivity of NO emissions to cross sections
The most recent use of the nonequilibrium model was to investigate the sensitivity of the calculated NO emissions to the cross sections for electron impact vibrational excitation of NO. Figure 9 shows three sets of integral cross sections for electron impact excitation of the $\nu' = 1$ level of NO. In addition to the cross sections [4] used in the “current” model above, there are excitation functions [12] scaled by $4\pi$ and theoretical calculations [13].

Figure 7. Calculated NO densities at 23:00 hours for the full calculation (●) and for the same calculations without the reaction $N_2(A^3\Sigma_u^+ + O \to NO + N(2D))$ (○) or without the enhancement of the $N(2D) + O_2$ rate (■).

Figure 8. Predicted emission rates for the NO $(\nu' \to \nu' - 1)$ emission due to electron impact (△), chemiluminescence (□), the sum of these two processes (●) and nightglow (○), and the rates deduced from the measurements (★). The nightglow values are magnified in the inset. Calculated values are given for a 5-kR aurora (lower values) and a 40-kR aurora (higher values).
Figure 9. Integral cross sections for electron impact vibrational excitation (0 → 1) of NO from measurements [4] (———), 4π-scaled excitation functions [12] (· · ·) and theory [13] (- - -).

Figure 10. Measured [8] NO fundamental infrared emissions (I) as a function of the vibrational level of NO. Also shown are current predictions for chemiluminescent reactions only (····), and for chemiluminescent reactions plus electron impact calculated for the current cross sections (● - ●), 4π-scaled excitation functions [12] (△––△) and theoretical calculations [13] (♦ - - - ♦).

The comparison of calculations (for a 40-kR aurora) with the rocket measurements is shown in figure 10. It can be seen that use of the scaled excitation functions improves the agreement for the (1 → 0) emission, while the use of the theoretical cross sections brings the calculated (1 → 0) emission into agreement with the measurements. However, for the (2 → 1) emission, the use of the scaled excitation functions and of the theoretical cross sections makes the calculated brightness too large [as compared to the higher fundamental lines, such as (3 → 2)]. A possible explanation is that the separation of the nightglow and auroral components [8] of the (1 → 0) emission was done incorrectly (possibly because the role of electron impact was not considered). Alternatively the relative rates of the chemiluminescent reactions are not correct, or the strength of the \( \nu' = 2 \) excitation is overestimated in the newer cross sections. The latter might be resolved by definitive low-energy cross-section measurements.

5. Conclusions
A nonequilibrium atmospheric model has been shown to predict NO densities and NO infrared emissions in good agreement with other models and with particular measurements. In each case the agreement with measurements is substantially improved when electron impact excitation is considered. At the height of the peak NO density around 110 km, inclusion of the \( \text{N}_2[\text{A}^3\Sigma_u^+] + \text{O} \) reaction increases the predicted NO density by a factor of \(~2\) at the equator and a factor of \(~3\) at high latitudes. An additional nonequilibrium calculation of processes driven by a limited-duration aurora gives predictions of NO infrared emissions in good agreement with rocket measurements of the spectral shape, except for the (1 → 0) emission. Addition of electron impact vibrational excitation of NO increases the (1 → 0) emission, thus improving the agreement of
the shape of the spectrum with the measurements. (There is an unexplained discrepancy for the absolute values.) Finally the calculations are repeated for more recent scaled excitation functions and theoretical integral cross sections for electron impact vibrational excitation of NO. These give, respectively, better and within-error agreement with the peak in the measured spectrum for the $(1 \rightarrow 0)$ emission, although both produce an increased discrepancy for the $(2 \rightarrow 1)$ emission. Improved experimental measurements of the NO electron impact vibrational cross sections are needed to investigate this discrepancy.

Acknowledgments

We thank the Australian Research Council for financial support of this research and the International Science Linkages programme of the Department of Innovation, Industry, Science and Research for funding international collaboration which led to the investigation of the sensitivity to NO electron impact cross sections.

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