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Analysis of the fluorescence emission from atmospheric nitrogen by electron excitation, and its application to fluorescence telescopes

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Abstract. A microscopic analysis of the processes involved in the fluorescence emission of nitrogen molecules induced by electronic collisions is carried out for a large range of incident energies (eV to GeV) and pressures (Pa to atmospheric conditions). The contribution of secondary electrons to that fluorescence is calculated by means of detailed Monte Carlo simulations. For this purpose, a novel analytical approximation of the energy spectrum of secondary electrons is used. The results of the simulations are shown to be useful for the interpretation of available experimental data. For instance, they account for the pressure dependence of the fluorescence observed in laboratory experiments. The conditions under which emitted fluorescence is proportional to deposited energy are also studied. Finally, these calculations provide an absolute value of the fluorescence yield consistent with available experimental data.

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1. **Introduction**

The origin and propagation of ultra-high-energy cosmic rays (>10^{18} eV) is at present one of the most important topics in astroparticle physics. The flux of cosmic rays at the highest energies is extremely small and therefore detectors with very large aperture have to be used. Basically two types of ground-based detectors are employed for this purpose: surface arrays and fluorescence detectors. Both aim at reconstructing the extensive air shower initiated by the primary cosmic ray. While surface arrays sample the shower tail, fluorescence detectors record the longitudinal development of the shower by means of wide-angle telescopes that observe the atmospheric fluorescence induced by the charged particles of the shower.

The fluorescence technique was used by the Fly’s Eye experiment [1] and later by its successor, HiRes [2]. The Telescope Array experiment [3] and the ASHRA project [4] also rely on this technique. Fluorescence telescopes are being employed very successfully by the...
Pierre Auger Observatory [5] operating simultaneously with a giant air shower array in a hybrid mode. On the other hand, the fluorescence traces of air showers could also be recorded from the top of the atmosphere in satellite experiments as JEM-EUSO [6], which proposes a super-wide field UV telescope for the detection of cosmic rays with energies above $10^{20}$ eV.

Fluorescence telescopes allow an accurate geometrical reconstruction of the shower axis, therefore providing the incoming direction of the primary cosmic ray. On the other hand, assuming that the fluorescence intensity is proportional to the EM energy released by the shower in the atmosphere, this technique provides a measure of the primary energy which is nearly model independent. Although this feature is an advantage, e.g. compared with surface arrays, the accuracy of this technique is at present limited by the uncertainty in the fluorescence yield, i.e. the calibration parameter that converts the number of fluorescence photons into absolute energy units. For instance in the Pierre Auger Observatory, the uncertainty in the fluorescence yield contributes 14% to the total systematic error in the energy calibration, which is at present 22% [7].

Measurements of the air-fluorescence yield have been carried out for many years [8]. Unfortunately, the accuracy of these pioneering studies is not enough for a precise calibration of fluorescence telescopes as demanded at present. In order to improve the accuracy in the fluorescence yield, several groups have been carrying out laboratory measurements since a few years ago [9]–[15]. Usually the experimental set-up consists of a beam of electrons that collides with an air target at known conditions (i.e. temperature, pressure, humidity, etc), generating fluorescence radiation that is then measured with an appropriate detection system. Electrons produced either in an accelerator or from a radioactive source are employed for this purpose. The goal of these experiments is manifold: measuring the absolute value of the fluorescence yield and determining of its dependence on wavelength and atmospheric parameters.

The wavelength spectrum of air fluorescence has been measured since long ago by many authors. The studied atmospheric parameters include pressure, composition and temperature. The dependence on pressure has been studied experimentally showing discrepancies between different authors that could be attributed, as discussed in this paper, to a different interpretation of the experimental data. The humidity dependence has been studied experimentally by several groups, reaching results that are in reasonable agreement with each other. Finally, recent measurements of temperature dependence indicate a possible non-negligible effect that should be taken into account in the energy reconstruction of cosmic rays. With regard to the absolute value of the fluorescence yield, a number of results are available. Most of them agree within about 20%. However, the comparison between modern experiments that claim higher accuracy (better than 10%) is not trivial because usually they have been performed under different experimental conditions, and thus the comparison requires a detailed analysis as will be shown in this paper. A review of the actual status of laboratory measurements with more references can be found in [16].

Progress in the theoretical understanding of the various processes leading to the air-fluorescence emission is also being made. The atmospheric fluorescence produced in a shower is due to the de-excitation of nitrogen molecules previously excited by the charged particles of the shower, mainly electrons. The study of the fluorescence emission caused by those electrons is the purpose of this work. The efficiency of high-energy electrons ($\gtrsim$ 1 keV) in the molecular excitation responsible for atmospheric fluorescence is very low. Instead, fluorescence is mainly generated by electrons with very low energy ejected in ionization processes. This important feature has to be understood well for an appropriate interpretation of the experimental results.
A model that accounts for the contribution of secondary electrons\(^2\) to the fluorescence has been recently published [17], allowing theoretical predictions of the total fluorescence yield as a function of the primary electron energy in a very wide interval (keV to GeV) [18].

Proportionality between fluorescence intensity and the energy deposited by a primary electron is usually assumed. Experimental results seem to support this assumption [11, 19, 20]. From a theoretical point of view, this proportionality is not obvious. The excitation of the fluorescence emission is strongly peaked at low energies (see figure 4(a)), in particular for the so-called 2P system of nitrogen, and therefore the ratio of fluorescence emission to deposited energy is strongly dependent on the spectrum of low-energy secondaries. In principle, this spectrum depends on both the primary energy and the distance from the primary interaction, and thus, this proportionality has to be demonstrated with a detailed analysis.

In this paper, the molecular processes involved in the generation of air-fluorescence radiation will be analyzed (section 2). Particular attention will be paid to the role of secondary electrons in the generation of fluorescence (section 3). Improvements of a procedure previously proposed for the evaluation of both the fluorescence light generated by secondaries and the deposited energy will be shown. For this purpose, a new analytical approximation for the energy spectrum of electrons ejected in ionization processes has been developed (section 4). As will be shown in this paper, a simple formula is able to predict the spectrum in a wide energy range of both primary and secondary electrons ranging from a few eV up to tens of GeV. Results on the fluorescence yield using this analytical spectrum will be shown in section 5. In particular, the dependence of the fluorescence yield on the primary electron energy and the size of the observation region related to the distance to the electron track (for a given pressure) will be calculated. In section 6, our results will be used for a discussion on the interpretation of some experimental data.

2. Molecular processes involved in the generation of air fluorescence

Electrons passing through the atmosphere lose energy by inelastic collision with air molecules. A small fraction of the deposited energy is emitted as UV fluorescence radiation in the spectral range 290–430 nm. Air fluorescence in this spectral range basically comes from the so-called 2P system of N\(_2\) and the 1N system of N\(_2^+\). Several features of the molecular excitation and de-excitation (radiative and collisional) will be recalled in this section. Theoretical predictions of relative intensities of the fluorescence spectrum will be compared with published experimental data. Finally, the relationship between fluorescence yield and molecular parameters will be discussed in section 2.4.

2.1. Molecular excitation by electron impact and radiative de-excitation

A schematic diagram of the main molecular levels of N\(_2\) and N\(_2^+\) involved in the fluorescence emission in our spectral range is shown in figure 1. Electron collisions excite (or ionize) molecular nitrogen in the ground state X \(^1\Sigma_g^+\) to upper electronic states of N\(_2\) and N\(_2^+\). As is well known from elementary molecular physics, each electronic state is split into vibrational levels \(v\). The set of electronic transitions \(v\rightleftharpoons v'\) between the level \(v\) of an upper electronic state

\(^2\) In the following, we give the name secondary electrons to those produced by ionization processes induced by a high-energy primary electron in a laboratory experiment.
Figure 1. Schematic diagram of the main nitrogen molecular levels involved in the air-fluorescence emission (red and blue arrows) following electron excitation (thin black arrows).

and the level \( v' \) of a lower state is named a band system. The second positive system (2P) results from the \( \text{N}_2 \ C^3\Pi_u \rightarrow B^3\Pi_g \) de-excitation, whereas the first negative system (1N) corresponds to the \( \text{N}_2^+ B^2\Sigma^+_u \rightarrow X^2\Sigma^+_g \) de-excitation after molecular ionization. Apart from the 1N and 2P systems, the weak bands of the \( \text{N}_2 \) Gaydon–Herman (GH) system have been observed in the fluorescence spectrum [13]. Data on wavelengths of the molecular bands of nitrogen are found in [21].

Upon passage of an electron, the probability of exciting an isolated nitrogen molecule from the ground state to an upper level \( v \) is given by the excitation cross section \( \sigma_v \). This cross section is proportional to the corresponding Franck–Condon factor \( q_{X \rightarrow v} \) (i.e. the overlapping integral between the vibrational wave functions of the ground state X and the upper level \( v \))\(^3\). Next the molecule may relax to a lower level \( v' \) by emitting a \( v-v' \) photon with an \( A_{vv'} \) probability per unit time (the Einstein coefficient). Therefore, in the absence of other processes apart from direct excitation and radiative de-excitation, the number of \( v-v' \) photons emitted per incident electron and unit path length is given by

\[
\varepsilon_{vv'} = N \sigma_v \frac{A_{vv'}}{A_v} = N \sigma_v B_{vv'} = N \sigma_{vv'},
\]

where \( N \) is the number of nitrogen molecules per unit volume, \( B_{vv'} \) is the branching ratio (i.e. the ratio between the partial \( A_{vv'} \) and total \( A_v = \sum_{v'} A_{vv'} \) radiative transition probabilities) and \( \sigma_{vv'} \) is the so-called optical cross section. As a result, the air-fluorescence intensity in the \( v-v' \) band is proportional to the \( q_{X \rightarrow v} B_{vv'} \) product. Values of the \( q_{X \rightarrow v} \) and \( B_{vv'} \) parameters for the nitrogen molecule are available in the literature [23, 24].

\(^3\) Franck–Condon principle [22].
2.2. Fluorescence quenching

Non-radiative molecular de-excitation by collision with other molecules of the medium (collisional quenching) becomes very important even at moderate pressures. The total de-excitation probability is the sum of the radiative and collisional transition probabilities $A_v + A_v^c$ and therefore the probability of $v \rightarrow v'$ photon emission diminishes as pressure grows. At a given temperature, $A_v^c(P)$ is proportional to the collision frequency and thus to the pressure $P$.

Defining $P_v^c$ as the characteristic pressure for which $A_v^c(P_v^c) = A_v$,

$$A_v^c(P) = A_v \frac{P}{P_v^c}. \quad (2)$$

This collisional quenching results in an additional pressure dependence of fluorescence emission. Replacing $A_v$ with $A_v + A_v^c$ in (1) results in the $1 + P/P_v^c$ Stern–Volmer correcting factor:

$$\varepsilon_{vv'}(P) = N\sigma_{vv'} \frac{1}{1 + P/P_v^c}. \quad (3)$$

Accordingly, the effective lifetime $\tau_v$ of the upper level is shortened with respect to the radiative one $\tau_v^r$ as

$$\frac{1}{\tau_v} = \frac{1}{\tau_v^r} \left( 1 + \frac{P}{P_v^c} \right). \quad (4)$$

Therefore, in a plot of $1/\tau_v$ versus $P$ (the Stern–Volmer plot), the slope provides the $P_v^c$ value. In principle, the characteristic pressure $P_v^c$ can also be obtained from the dependence of the fluorescence intensity on pressure as given by (3). However, equation (3) only accounts for light from primary interactions, whereas the observed fluorescence in practical conditions includes the contribution from secondary electrons as will be explained below. Although the quenching correction in (3) is valid, in a laboratory experiment, losses of fluorescence due to secondary electrons escaping the field of view can induce a further pressure dependence on the total measured fluorescence (see section 5.1 for details). As will be shown in section 6.1, neglecting this effect can cause an overestimation of $P_v^c$ values.

All atomic and molecular components of atmospheric air might act as quenchers for excited nitrogen (i.e. nitrogen, oxygen, water vapor and pollutants); in particular, oxygen molecules are known to be very efficient for collisional de-excitation of air fluorescence. In general, the characteristic pressure for a mixture of gases follows the law

$$\frac{1}{P'} = \sum_i f_i \frac{1}{P_i'}, \quad (5)$$

where $f_i$ is the fraction of molecules of type $i$ (including nitrogen), and the partial characteristic pressures $P_i'$ can be expressed in terms of elementary molecular parameters as

$$P_i' = \frac{kT}{\tau_i \sigma_{Ni} \bar{v}_{Ni}}, \quad \bar{v}_{Ni} = \sqrt{\frac{8kT}{\pi \mu_{Ni}}}. \quad (6)$$

In the above relations $k$ is Boltzmann’s constant, $T$ is the temperature of the gas, $\sigma_{Ni}$ is the cross section for collisional de-excitation of an excited nitrogen molecule by molecules of type $i$, and $\bar{v}_{Ni}$ and $\mu_{Ni}$ are the relative velocity and reduced mass of the two-body system $N–i$, respectively.
Figure 2. As predicted by (8), experimental relative intensities [13] of the 2P system turn out to be proportional to the $q_{X \rightarrow v} B_{vv'} P_v'$ factor. This confirms the applicability of the Franck–Condon principle for electron excitation of these bands (different upper $v$ levels being involved) and shows good agreement with previously known branching ratios and the Franck–Condon factors [23]. The necessary $P_v'$ values were taken from [13].

Equation (6) shows a net $\sqrt{T}$ growing behavior of the characteristic pressure. However, the collisional cross section is expected to be a function of the kinetic energy of the colliding particles, and so $\sigma_{ni}$ is also $T$ dependent. Although data on this temperature dependence is very scarce, in many cases a $\sigma_{ni} \propto T^\alpha$ power law can be assumed [25], where $\alpha$ might take different values according to the dominating type of interaction for each quencher. Recent measurements [26, 27] confirm this additional temperature dependence of the fluorescence emission.

2.3. Theoretical predictions on relative intensities

From the above molecular properties of the air-fluorescence emission, some predictions on relative intensities of the fluorescence spectrum can be inferred. For a given band system, the relative intensities $I_{vv'}$ (with respect to a reference one; e.g. $I_{00}$) are given by

$$\frac{I_{vv'}}{I_{00}} = \frac{q_{X \rightarrow v} B_{vv'} P_v'}{q_{X \rightarrow 0} B_{00}} \frac{1 + P/P_0'}{1 + P/P_v'},$$

which at high pressures ($P \gg P_v'$) becomes

$$\frac{I_{vv'}}{I_{00}} \approx \frac{q_{X \rightarrow v} B_{vv'} P_v'}{q_{X \rightarrow 0} B_{00} P_0'},$$

Therefore, in the absence of other population mechanisms, relative intensities of a band system at high pressure are expected to be proportional to $q_{X \rightarrow v} B_{vv'} P_v'$ as pointed out in [17]. This property can be easily verified using available experimental data. In figure 2, accurate measurements of the relative intensities of the 2P system bands carried out by the AIRFLY collaboration are plotted against $q_{X \rightarrow v} B_{vv'} P_v'$, using the tabulated values of $q_{X \rightarrow v}$ and $B_{vv'}$. 

from [23] and the experimental characteristic pressures reported by AIRFLY [13]. In the resulting plot a remarkable linearity is found, in particular for intense bands. Note that this analysis includes all molecular transitions no matter what their upper levels $v$ are. This result provides an experimental verification of the applicability of the Franck–Condon principle for the electron excitation of the upper levels of the 2P system of nitrogen.

It must be noted that the experimental values used here correspond to the total observed fluorescence, whereas expressions (7) and (8) were obtained considering only primary processes. Nevertheless, as will be shown in sections 3 and 5, the fluorescence contributions from primary and secondary electrons present the same wavelength dependency (neglecting any energy dependence of the Franck–Condon factors).

An important consequence of equation (7) is the possibility of obtaining results for different bands from a given one. For that reason, the main results in sections 5 and 6 will be given only for (0, 0) bands of 1N and 2P systems.

2.4. Definition of the fluorescence yield

In the literature, several parameters have been used to measure the amount of fluorescence light emitted by an electron that deposits energy in the atmosphere. A detailed description of the various magnitudes can be found in [16]. The number of photons emitted per electron and unit path length is a valuable parameter that is used extensively. Neglecting the contribution of secondary electrons, equation (3) relates this parameter $\varepsilon_{vv'}$ with the optical cross section including the Stern–Volmer factor that accounts for quenching effects. However, as will be discussed in section 3, secondary electrons ejected in ionization processes generate a significant amount of fluorescence not included in (3). It will be shown below that an effective optical cross section $\sigma_{vv'}^{\text{eff}}$ that accounts for this contribution can be defined. This effective cross section turns out to be dependent on the gas pressure and the size of the observation region. Therefore, in practice, equation (3) becomes

$$
\varepsilon_{vv'} = N\sigma_{vv'}^{\text{eff}}(E, P) \frac{1}{1 + P / P_v'},
$$

(9)

where $\varepsilon$ represents now the total experimentally observed fluorescence, produced by primary as well as secondary electrons.

Following an already common usage, in this paper we will give the name fluorescence yield $Y_{vv'}$ to the number of photons $v-v'$ per unit energy deposited in the medium. The relationship between both parameters is

$$
Y_{vv'} = \frac{\varepsilon_{vv'}}{(dE/dx)_{\text{dep}}},
$$

(10)

where $(dE/dx)_{\text{dep}}$ is the energy deposited per primary electron and unit path length. As will be discussed later, the application of this relationship requires that both the emitted photons and the deposited energy correspond to the same air volume (see figure 3).

3. Role of secondary electrons in the generation of air fluorescence

As mentioned above, most of the fluorescence emitted by an extensive air shower is generated by secondary electrons of very low energy. This feature of the air fluorescence has relevant
Figure 3. Secondary electrons moving away from the primary interaction point cause fluorescence emission and energy deposition outside the primary electron trajectory. At large primary energies, the contribution from a small number of high-energy secondaries depends on the geometry of the interaction region.

implications in the interpretation of experimental data on fluorescence yields carried out in laboratories.

The leading role of secondary electrons becomes apparent when comparing the behavior of the nitrogen fluorescence at low and high pressure. The 2P system fluorescence excited by low-energy electrons ($E \lesssim 1$ keV) at very low pressure ($\lesssim 0.1$ Pa) [28]–[30] shows a fast $E^{-2}$ dependence, consistent with the optically forbidden nature of the transition $X^1\Sigma_g^+ \rightarrow C^3\Pi_u$.

For comparison, the excitation of the 1N system of $N^+_2$ ($X^1\Sigma_g^+ \rightarrow B^2\Sigma_u^+$) exhibits a much slower $(\log E)/E$ dependence. A simple extrapolation of these excitation laws (see figure 4(a)) predicts that the 2P fluorescence should be negligible at energies larger than about 10 keV. However, it is well known that fluorescence in both air and pure nitrogen at pressures over a few hPa is dominated by the 2P system [31].

The large amount of 2P fluorescence observed at high pressure arises from excitations by low-energy secondary electrons ejected in ionization processes. Although this explanation was first suggested by Bunner [32], it has not been quantitatively verified until very recently [17] by means of a detailed simulation that takes into account all the related molecular processes. Since secondary electrons travel a certain air thickness before stopping, their fluorescence contribution depends on the pressure and the geometrical features of the observation region. Nevertheless, as shown in [17], even a very simple model that assumes that the interaction volume is a sphere can account for several experimental results on the 2P fluorescence of nitrogen, using the radius of the sphere as a single parameter for the size of the observation region.

The above-mentioned simple model is briefly reviewed below (3.1) and the detailed simulation is described later (section 3.2). Although in [17] this analysis is carried out for the 2P system, the procedure can be also applied to the 1N system as shown in [18].

3.1. Dependence on the pressure and size of observation region: a simple model

For simplicity, let us assume that a primary electron has interacted at the center of an air sphere of radius $R$ (see figure 3). Secondaries ejected at this point generate an amount of fluorescence
inside this sphere that can be evaluated under several approximations. If the mean free path of these secondaries is small compared with $R$, they will contribute significantly to further excitation or ionization of other molecules. Note that, due to the strong energy dependence of inelastic cross sections (figure 4(a)), an interaction region might be thin for the primary electron but thick for lower-energy secondary ones.

As mentioned above, neglecting collisional de-excitation, the number of $v-v'$ photons per unit path length produced by primary collisions is given by the optical cross section (1). For each primary incident electron, the number of secondaries generated per unit length is $N_{\text{air}} \sigma_{\text{ion}}(E)$, where $N_{\text{air}}$ is the molecular density of the medium and $\sigma_{\text{ion}}(E)$ is the ionization cross section\(^4\). The fraction of these secondaries undergoing at least one inelastic interaction before escaping is given by $(1 - e^{-N_{\text{air}} \langle \sigma_{\text{inel}} \rangle R})$, where $\langle \sigma_{\text{inel}} \rangle$ is the mean value of the inelastic cross section for the energy distribution of secondary electrons. The fraction of these interacting secondary electrons that generates $v-v'$ photons can be estimated as $\langle N\sigma_{vv'}/N_{\text{air}}\sigma_{\text{inel}} \rangle$, that is, $N/N_{\text{air}}$ times the mean value of the $\sigma_{vv'}(E)/\sigma_{\text{inel}}(E)$ ratio under the energy distribution of secondary electrons.

Thus, neglecting collisional quenching, the total number of fluorescence photons per unit path length including secondary processes can be finally estimated as

$$\varepsilon_{vv'}^{0}(E) = N\sigma_{vv'}(E) + N\langle \sigma_{vv'}/\sigma_{\text{inel}} \rangle (1 - e^{-N_{\text{air}} \langle \sigma_{\text{inel}} \rangle R}) \sigma_{\text{ion}}(E).$$

(11)

An ‘effective cross section’ that includes the fluorescence contribution of secondaries can be defined by $\varepsilon_{vv'}^{0}(E) = N\sigma_{vv'}^{\text{eff}}(E)$, i.e. the cross section that would give rise to the observed

\(^4\) Strictly speaking $\sigma_{\text{ion}}$ is the so-called ‘gross ionization cross section’ [33, 34], which also includes processes generating more than one electron although their contribution is small. Oxygen cross sections are approximated here by nitrogen ones.
fluorescence intensity by direct excitation, and is therefore given in this approximation by the expression

\[ \sigma_{\text{eff}}(E) = \sigma_{v'v}(E) + \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle (1 - e^{-N_{\text{air}}(\sigma_{\text{inel}})R}) \sigma_{\text{ion}}(E), \]

(12)

where primary and secondary contributions can be clearly distinguished. From the above result, some interesting consequences can be drawn in the low- and high-pressure limits.

At very low pressures, the contribution of secondary electrons can be neglected since the interaction is thin for secondaries, i.e. \( \langle \sigma_{\text{inel}} \rangle N_{\text{air}} R \ll 1 \), and therefore the fluorescence intensity becomes proportional to \( \sigma_{v'v}(E) \). At very large pressures, the interaction region is thick for secondaries, i.e. \( \langle \sigma_{\text{inel}} \rangle N_{\text{air}} R \gg 1 \), and the exponential term saturates. In this case, for the 2P system at high energy,

\[ \sigma_{\text{eff}}(E) \approx \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle \sigma_{\text{ion}}(E), \]

(13)

and therefore, 2P fluorescence is mainly generated by secondaries with \( \epsilon_{v'v}(E) \) being proportional to \( \sigma_{\text{ion}}(E) \). For the 1N system, the cross section \( \sigma_{v'v}(E) \) is proportional to \( \sigma_{\text{ion}}(E) \) and the contribution of secondary electrons only represents a (non-negligible) correction to the one generated by direct excitation [18]. Note that the results are almost independent of the geometry in those two limit situations.

Figure 4(b) illustrates the energy dependence of emitted fluorescence in those two limit situations and several other intermediate ones, by comparing the predictions of the model with the experimental values obtained in different \( N_{\text{air}} \cdot R \) conditions. For this purpose, the necessary \( \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle \) values were calculated in [17] and are almost energy independent above a few tens of eV. This is a nontrivial calculation, as described in the next sections.

From the above discussion, the characteristic thickness \( N_{\text{air}} \cdot R \) that separates the two limit situations is given by \( 1/\langle \sigma_{\text{inel}} \rangle \), where the average cross section\(^6\) \( \langle \sigma_{\text{inel}} \rangle = 5.6 \pm 0.4 a_0^2 \) is almost independent of the primary electron energy. For practical purposes, it is more convenient to express the thickness as a \( P \cdot R \) product, i.e. in terms of pressure \( P \) instead of density of molecules. The characteristic thickness in \( P \cdot R \) units equals \( 0.26 \pm 0.02 \text{hPa cm}^{-1} \) and gives a measure of the range of most secondary electrons (low-energy ones) in air.

The application of (11) for the calculation of the fluorescence photons per unit path length requires the evaluation of \( \langle \sigma_{\text{inel}} \rangle \) and \( \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle \) and therefore an important ingredient of the model is the energy distribution of secondary electrons generated by a primary one at a given energy. Assuming that this spectrum is known (see section 4), \( \langle \sigma_{\text{inel}} \rangle \) can be easily calculated using available inelastic cross sections in air. Evaluation of \( \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle \) as a simple average of the \( \sigma_{v'v}(E)/\sigma_{\text{inel}}(E) \) quotient for the energy distribution of secondary electrons would only take into account the first interaction of each secondary. Although high-energy secondaries are scarce, their contribution should not be ignored as they can be involved in several further excitation and ionization processes. The analytical evaluation of \( \langle \sigma_{v'v}/\sigma_{\text{inel}} \rangle \) taking into account further processes of high-energy secondaries would require a knowledge of their energy distribution after successive interactions, which is a nontrivial problem. For this purpose, a Monte Carlo simulation is very useful because it also allows fine details of the excitation and ionization processes to be included, as well as the detailed spatial distribution of the energy deposition and fluorescence emission to be obtained, for any geometry or experimental condition.

\(^5\) This result is not applicable to the 2P system at high energy since \( \sigma_{\text{ion}}(E) \) is larger than \( \sigma_{v'v}(E) \) by several orders of magnitude and therefore, even at pressures as low as 0.1 Pa, direct excitation is not dominant.

\(^6\) In this paper, cross sections are expressed in squared Bohr radius units.
3.2. Detailed Monte Carlo analysis of the processes involved

A simple C++ code has been developed which allowed us to follow the trajectory of a primary electron with arbitrary initial velocity as it interacts with a selected nitrogen pressure inside an interaction region of chosen geometry. As described below, the different molecular processes involved were characterized by the appropriate parameters and the history of primary and secondary electrons could be followed down to 11 eV energy. Below this energy, electrons are not able to ionize or to generate light emission of interest.

For each electron inside the simulated interaction region, the trajectory was a succession of ‘free flight’ steps randomly generated according to a Poisson distribution, using as the average step size the \((N\sigma)^{-1}\) mean free path obtained from the gas density and total cross section \(\sigma(E) = \sigma_{\text{el}}(E) + \sigma_{\text{ion}}(E) + \sigma_{\text{exc}}(E)\) including elastic, ionization and excitation processes. After each step the nature of the interaction was randomly decided according to the ratio of those cross sections.

When the selected process is an elastic interaction, the electron energy does not change but a new direction is randomly assigned, according to the differential elastic cross sections [35]. The final results turned out to be weakly dependent on the fine details of these differential cross sections, and so approximate expressions were used for them in order to save computing time.

For inelastic processes, the same approximate differential cross sections were also used, but in addition to the change in direction, the electron energy was reduced as described below.

The three inelastic processes considered were:

1. **Excitation (without ionization)**. In this case, the electron energy is reduced by an amount equal to the average excitation energy \(\langle E_{\text{exc}} \rangle\), which is annotated as deposited in the gas. In addition, a number of \(\sigma_{337}(E)/\sigma_{\text{exc}}(E)\) 337 nm photons are emitted.

2. **Ionization**. In this case, a number of \(\sigma_{391}(E)/\sigma_{\text{ion}}(E)\) 391 nm photons are emitted and a secondary electron is generated with random energy according to the corresponding distribution (see equation (16) in section 4.1). In this way, an additional electron is incorporated into the simulation. This energy is discounted from the electron together with the ionization potential and the average excitation energy of the ion \(\langle E_{\text{exc}}^{\text{ion}} \rangle\), which are annotated as deposited in the gas. With a probability corresponding to the cross section for K-shell ionization, 410 eV are also discounted from the original electron and annotated as x-ray emission.

3. **Bremsstrahlung**. In this case, the loss of energy is estimated as 3% of the electron energy [36] and annotated as gamma-ray emission. Bremsstrahlung effects become significant only at very large electron energies.

It should be noted that both bremsstrahlung and K-shell ionization are rare processes when compared with the (several orders of magnitude larger) ionization cross section. For that reason, they are important for energy-balance purposes but not for fluorescence-photon-counting purposes; hence the approximate treatment is used here for them.

The simulation is initiated with a primary electron located at a certain position moving in a chosen direction with a given energy. This electron follows a track as described above until it stops or exits some predefined geometrical limits. Secondary electrons are incorporated into the simulation and treated in the same way as they are generated in ionization processes. During the simulation, the deposited energy and fluorescence emission are evaluated as explained above.
The typical number of events to achieve reasonable statistics has been about $10^6$. In this way results for an arbitrary experimental geometry can be obtained.

Alternatively, in order to obtain results for a generic spherical geometry like the one described in section 3.1, secondaries were generated at the center of the sphere following the energy distribution corresponding to a given primary energy. When operating in this mode the interesting result of the simulation is the total number of $v\rightarrow v'$ photons emitted per event (i.e. per secondary electron) inside the sphere. This magnitude $\alpha_{vv'}$, described in section 5.1, will be very useful for making general predictions on the fluorescence contribution of secondary electrons. Note that the simulations were carried out for pure nitrogen. For that reason, an $N/N_{air}$ correcting factor will be included when using these results for obtaining fluorescence yield in air.

Necessary ingredients for the simulation are the cross sections of all the processes and the average excitation energies. The cross sections used in this work are shown in figure 4(a). They have been compiled from the bibliography as follows. Values for $\sigma_{\text{opt}}$ optical cross section were taken from [37] up to 250 eV. At larger energies, a Born–Bethe extrapolation fitting these and many other values (see [18] for more details) was adopted. Gross ionization cross section values were taken from [34] up to 1 keV energies. At larger energies, these values and those from [38] were used to fit a Born–Bethe extrapolation with the same slope used for the $\sigma_{\text{opt}}$ cross section fit. Nitrogen K-shell ionization cross section was taken from [39] in the 410 keV–100 MeV range, and extrapolated as proportional to the gross ionization one at larger energies. Values for $\sigma_{337}$ optical cross section were an average of those from [29] and [30]. Values for bremsstrahlung cross section were taken from [36] and for numerical purposes they were approximated as a $2 \times 10^{-5}$ fraction of the gross ionization cross section. The total excitation cross section of $N_2$ was taken from [40] in the 0–40 eV region and from [35] for energies up to 1 keV and extrapolated as proportional to the ionization cross section at larger energies. Total cross section values (including elastic and inelastic processes) were taken from [41] up to 1 keV energies and Born–Bethe extrapolated at larger ones. The mean excitation energy of $N_2$ levels was estimated as $\langle E_{\text{exc}} \rangle = 8.5$ eV by averaging the available data on energy levels weighted with their corresponding excitation cross sections taken from [42]–[44]. In a similar way, the average excitation energy of $N_2^+$ levels was estimated in $\langle E_{\text{exc}} \rangle = 1.3$ eV from the data in [45].

While all those values are relatively well known, they present uncertainties ranging from about 5% in the 10 eV–1 keV region to 20% at 10 keV and larger energies. While the results on spatial distributions and energy dependencies can be considered reliable, relative values of 1N and 2P fluorescence are probably accurate only within 10% and absolute values only within 20%.

An additional critical ingredient in this treatment is the energy distribution of secondary electrons. As this deserves special attention it will be discussed in the next section.

4. Energy spectrum of secondary electrons

4.1. Experimental and theoretical data in different energy ranges: an analytical formula

The energy distribution of secondary electrons originating from ionization of $N_2$ molecules has been experimentally determined by several authors for primary electrons with kinetic energies up to several keV [46]–[48], and several expressions have been proposed that fit the resulting
distributions. A very simple one originally proposed by Opal et al [46] was
\[
\frac{d\sigma_{\text{ion}}}{dE_s} = \begin{cases} 
\frac{C(E_p)}{E_s^2 + w^2}, & \text{for } E_s \leq (E_p - I)/2, \\
0, & \text{for } E_s \geq (E_p - I)/2,
\end{cases}
\]
(14)
where \( E_p \) is the kinetic energy of the primary electron\(^7 \), \( E_s \) is that for the secondary one, \( I = 15.6 \text{ eV} \) is the \( \text{N}_2 \) ionization potential, and the parameter \( w = 11.4 \text{ eV} \) for nitrogen. The normalization constant \( C(E_p) \) must be chosen in such a way that the integrated distribution reproduces the well-known \( \sigma_{\text{ion}} \) values.

Due to electron indistinguishability, the identification of primary and secondary electrons after an ionization process is not possible, and the standard procedure consists of labelling as secondary the one with the lowest kinetic energy. For that reason the maximum allowed value for \( E_s \) is \((E_p - I)/2\).

A careful calculation of the \( d\sigma_{\text{ion}}/dE_s \) energy distribution of secondary electrons at large primary energies for which no experimental data are available is complicated, and has been described by Seltzer [36, 50]. Numerical results for any atom are available in EEDL databases [36]\(^8 \), but include the density correction only at a fixed pressure. Next we will justify that expression (14), used in our previous studies [17, 18] for air fluorescence, is in general not able to describe properly the energy spectrum of secondaries at large \( E_p \) values.

Firstly, the energy distribution of high-energy secondary electrons (i.e. ‘delta rays’) follows that of the well-known Möller \( e^-e^- \) scattering. However, the Möller energy distribution could only be approximated by (14) if the normalization constant \( C(E_p) \) takes the \( 4\pi Z \text{Ry}^2/E_p' \) value [49], where \( Z = 14 \) for nitrogen, \( \text{Ry} \) is the Rydberg constant, \( E_p' = \frac{1}{2}m_e\beta^2c^2 \) and \( m_e \) is the electron rest mass\(^9 \). Since the \( C(E_p) \) constant has to be fixed according to the independently well-known \( \sigma_{\text{ion}} \) values, in general both conditions cannot be fulfilled simultaneously.

Secondly, in the GeV energy region, the total cross section depends on pressure through the density correction, low-energy secondaries (mostly arising from distant collisions) being more affected than large-energy ones (mostly arising from close collisions). As a consequence, the average energy of secondary electrons and the shape of their distribution must also be pressure dependent. None of these effects can be described by the fixed form (14), for which the average energy of secondaries for \( E_p \gg I \) is
\[
\langle E_s \rangle = \frac{\int E_s (d\sigma_{\text{ion}}/dE_s) dE_s}{\int (d\sigma_{\text{ion}}/dE_s) dE_s} \approx \frac{2w}{\pi} \ln \frac{E_p}{2w},
\]
(15)
which is independent of the normalization constant and hence of the total cross section and pressure conditions. Also, as mentioned above, ionization cross sections and energy distribution of secondaries are only available from EEDL databases for a given density correction at a fixed pressure, whereas for our applications, data at different pressures will be necessary. These are obtained by including the density correction, as described in [18], as a function of pressure.

In this way, for a valid family of energy distributions for secondary electrons, the requirements are: good agreement with the Opal expression at low primary energies where this is very reliable, good agreement with EEDL distributions when density correction is included for

\(^7\) In this section, the energy of the primary electron will be named \( E_p \) to stress the difference from the energy of the secondaries. In fact, this \( E_p \) energy is usually equivalent to that labelled \( E \) in other sections.

\(^8\) EEDL databases are included in GEANT4 packages or available at http://www.nds.iaea.org/epdl97/.

\(^9\) For energies lower than 10 keV, \( E_p' \) nearly equals the kinetic energy of the electron.
the pressure for which they are calculated and good agreement at all pressures with δ-ray energy distributions for relativistic primary–secondary energies. As described in the next section, the excitation and ionization cross sections, when combined with the average excitation energies and average energy of secondary electrons \( \langle E_s \rangle \), must reproduce the well-known stopping powers, and this results therefore in a very important additional requirement for the desired distributions. 

An analytical expression for \( d\sigma_{\text{ion}} / dE_s \) was desirable for the practical purpose of being implemented in our Monte Carlo calculations, and this was achieved as follows. For \( E_p \) values up to 4.5 keV, we use the Opal expression (14) that agrees very well with experimental as well as EEDL values. At larger energies, the following new expression was used:

\[
\frac{d\sigma_{\text{ion}}}{dE_s} = \begin{cases} 
4\pi Z Ry^2 \frac{1 + C(E_p) e^{-E_s/E_k}}{E_p' E_s^2 + w^2}, & \text{for } E_s \leq (E_p - I)/2, \\
0, & \text{for } E_s \geq (E_p - I)/2,
\end{cases}
\]  

(16)

where the normalization constant \( C(E_p) \) is fixed as in the Opal expression. 

The two expressions (14) and (16) coincide at 4.5 keV where one finds \( C(E_p) = 0 \) in (16). Expression (16) is inspired by the Seltzer procedure [36, 50] of distinguishing close and distant collisions, where the \( E_k \) parameter acts as the boundary between the two kinds of processes. In fact, the \( E_k = 77 \) eV value used in this work was adopted as an average for the binding energy of \( \text{N}_2 \) shells, which ranges from 410 eV (K-shell) down to 15.6 eV (valence electrons). In this paper, this analytical energy spectrum, which as shown below is applicable up to very high energies, will be named the ‘extended’ Opal formula. 

At secondary energies well above \( E_k \), the exponential vanishes and so expression (16) reduces to the well-known energy distribution of δ-rays arising from the Möller e\(^-\)–e\(^-\) scattering, whereas at secondary energies below \( E_k \), expression (16) behaves like (14) with a normalization constant depending on pressure through the effect of density correction on \( \sigma_{\text{ion}} \). 

An efficient random generation of energy values according to distribution (16) was numerically carried out in two steps. Firstly, a random value distributed as \( 1/(E_s^2 + w^2) \) was calculated by the usual procedures, and secondly, the rejection procedure comparing the very similar \( 1/(E_s^2 + w^2) \) and (16) distributions was used to obtain the desired value distributed as (16). 

The secondary energy distributions (16) are illustrated in figure 5 for 1 MeV and 1 GeV primary electron energies. For the \( E_p = 1 \) MeV case, for which the density correction is negligible, expression (16) fits well the EEDL shape in the whole secondary electron energy range. At \( E_p = 1 \) GeV, our analytical formula illustrates the change in the low-energy region of the distributions due to the pressure effect. Nevertheless, expression (16) fits the numerical values in EEDL databases for all the primary electron energies when including the density correction corresponding to the pressure for which they are calculated. 

The resulting mean value of the energy distribution of secondaries \( \langle E_s \rangle \), calculated as (15), is shown in figure 6(a) versus \( E \) for both the Opal (14) and our extended Opal (16) distributions. As mentioned above, the simple Opal formula gives a fixed energy dependence of \( \langle E_s \rangle \) at high energies, while our extended Opal formula provides the proper pressure-dependent shape accounting for the density correction.
Figure 5. Comparison of the energy spectrum of the secondary electrons from several analytical approximations with those values from the EEDL database (see footnote 8) at 1 MeV (a) and 1 GeV (b) primary electron energies. At large energies, both EEDL and the extended Opal formula proposed in this work approximate correctly the Möller distribution for \( \delta \)-rays, whereas the Opal expression (14) overestimates it by up to a factor of 2 at large energies (b). EEDL values include density correction at a large pressure, whereas the Opal and the extended Opal values do not in these figures, in order to illustrate the change in shape this causes in the energy distributions.

4.2. Total energy losses: comparison with the Bethe–Bloch theory

The analytical energy spectrum (16) contains only one adjustable parameter \( C(E_p) \), which is fixed by the normalization condition \( \int dE_s (d\sigma_{ion}/dE_s) = \sigma_{ion} \). However, the energy spectrum also has to be able to predict the Bethe–Bloch stopping power properly, although no degree of freedom remains for imposing this condition. As will be shown next, the energy spectrum (16) with the normalization constant calculated as mentioned above predicts the stopping power of the primary electron very accurately. This fact is a powerful consistency check of the proposed analytical spectrum.

The stopping power is closely related to the mean value of the energy distribution of secondary electrons. In fact, the energy loss per unit path length must result from the number of interactions per unit path length multiplied by the average value of the energy loss per interaction [36, 51]. In the case of pure molecular excitation, the average energy loss is \( \langle E_{exc} \rangle \), whereas in the case of ionization the loss amounts to the average excitation energy of the ion \( \langle E_{ion}^{exc} \rangle \) plus the ionization energy and the average value of the kinetic energy of the ejected electron. For air, assuming \( N_{air} \) molecules per unit volume,

\[
\frac{1}{N_{air}} \frac{dE}{dx} = \langle E_{exc} \rangle \sigma_{exc}(E) + \left[ \langle E_{ion}^{exc} \rangle + \langle E_s \rangle + I \right] \sigma_{ion}(E),
\]

which can also be expressed as

\[
\frac{dE}{dx} = N_{air} \left[ \langle E_{dep}^0 \rangle + \langle E_s \rangle \right] \sigma_{ion}(E)
\]
by separating the $\langle E_s \rangle$ energy carried out by secondaries from the $\langle E_{dep}^0 \rangle$ locally deposited one,

$$\langle E_{dep}^0 \rangle = \langle E_{exc} \rangle \frac{\sigma_{exc}}{\sigma_{ion}} + \langle E_{ion}^{exc} \rangle + I,$$

where a $410 \sigma_K/\sigma_{ion} \approx 5$ eV contribution must also be included arising from K-shell ionization at energies above 410 eV.

The result of (17) has been evaluated using the $\langle E_{exc} \rangle$ and $\langle E_{ion}^{exc} \rangle$ values\(^\text{10}\) (see section 3.2) and the $\langle E_s \rangle$ predicted by the extended Opal formula.

In figure 6(b), the result for the extended Opal formula including the density correction is compared with the Bethe–Bloch energy loss [52]. The very good agreement shown in this figure is a validity test of (16). It can be easily checked that the values in figure 6(a) from the simple Opal formula (14) are inconsistent with those of the Bethe–Bloch theory. Therefore, the use of the extended Opal formula proposed in this work improves significantly our theoretical predictions on the fluorescence yield, as shown below, in comparison with others previously reported [18].

5. Predictions of the model

In this section, results of the theoretical approaches presented in section 3 will be shown. The fluorescence intensity generated per secondary electron as a function of $E$, $P$ and the size of the interaction region will be evaluated in section 5.1. We will show that even the simple model of section 3.1 is able to make interesting predictions although more accurate results can be

\(^{10}\) Average excitation energies for air are very similar to those of pure nitrogen because, as already indicated, the total excitation and ionization cross sections of nitrogen and oxygen molecules are very close to each other.
achieved from the simulation described in section 3.2. Results on the energy deposited per unit path length by a primary electron will be calculated in section 5.2. Finally, the relationship between fluorescence light and deposited energy will be studied in section 5.3. In addition, theoretical estimates on the fluorescence yield will be presented.

5.1. Pressure and spatial dependency of the fluorescence contribution from secondary electrons: the \( \alpha \) parameters

As defined above, \( \alpha_{vv'} \) is the average number of \( v-v' \) photons produced in pure nitrogen inside a given region per first-generation secondary electron, that is, per secondary arising directly from ionizations caused by the primary electron. The value of \( \alpha_{vv'} \) can be evaluated as a function of \( E, P \) and the geometrical features of the interaction region using our simulation. This parameter allows the determination of the effective optical cross section by combining the \( N\sigma_{vv'} \) contribution from primary interactions with the \( N/N_{air}\alpha_{vv'} \) one from each of the \( N_{air}\sigma_{ion} \) first-generation secondaries. This effective cross section,

\[
\alpha_{vv'}^{\text{eff}}(E, P) = \alpha_{vv'}(E) + \alpha_{vv'}(E, P) \cdot \sigma_{ion}(E),
\]

allows the determination of the total number of fluorescence photons per unit path length from (9). Comparison of (20) and (12) shows that the \( \alpha_{vv'} \) values were given within the simple model (section 3.1) by

\[
\alpha_{vv'} = \langle \sigma_{vv'}/\sigma_{\text{mel}} \rangle \left( 1 - e^{-N_{air}R(\sigma_{\text{mel}})} \right).
\]

If we neglect the pressure dependence in the ionization cross section due to the density correction, for the spherical geometry \( \alpha_{vv'} \) is a function of the \( P \cdot R \) product [17], that is, a function of the thickness of the interaction region.

The Monte Carlo results for the 2P(0, 0) band (337 nm) and the 1N(0, 0) band (391 nm) are shown in figure 7 as a function of primary energy (up to \( E = 20 \text{ GeV} \)) and for \( P \cdot R \) values in the interval \( 0.1-10^3 \text{ hPa} \times \text{cm} \). The results shown in this figure have been obtained neglecting the density correction and therefore they are valid at low pressure. From our simulations we have found that the effect of density correction is to keep nearly constant the \( \alpha_{vv'} \) parameter at energies above a minimum pressure-dependent value \( E_{\text{min}}(\text{GeV}) = 3.2/\sqrt{P(\text{hPa})} \). Therefore the results of figure 7 are valid at atmospheric pressure for energies below 0.1 GeV, whereas the density effect on the \( \alpha_{vv'} \) parameters at 10 GeV amounts to about 10% (25%) for the 2P (1N) system assuming an \( R \) value in the range of a few centimeters. Dotted lines in figure 7 represent the result for a very large medium (\( R \rightarrow \infty \)), which shows large fluctuations. Note that accurate results for a very large medium would need a huge number of simulated events.

The \( P \cdot R \) dependence of \( \alpha_{vv'} \) parameters can be better appreciated in figure 8. At low values of the \( P \cdot R \) product this dependence is linear, as then secondary electrons can escape the interaction region, and the amount of deposited energy and light is proportional to this thickness. At larger values of the \( P \cdot R \) product, \( \alpha_{vv'} \) values become very weakly dependent on the thickness as then the interaction region is thick for the secondaries and almost none escapes. For both the 1N and 2P band systems, the \( P \cdot R \) value that separates both regimes is \( \langle \sigma_{\text{mel}} \rangle^{-1} \) corresponding to 0.26 hPa \( \times \text{cm} \) as predicted by the simple model of section 3.1. This value corresponds to the average range of secondary electrons in nitrogen (or air).

It must be noted that, at large energies, \( \alpha_{vv'} \) values present a small logarithmic growth even for \( P \cdot R \) values well above the \( \langle \sigma_{\text{mel}} \rangle^{-1} \) limit. This is due to the contribution of a very small
Figure 7. Dependence of $\alpha_{00}$ values (in photons per secondary electron) on the energy of the primary electron for several values of $P \cdot R$ (i.e. thickness of the interaction region). Monte Carlo results for the 2P (a) and 1N (b) systems are shown for spherical geometry. These results do not include density correction. Values at higher pressures can be obtained as described in the text.

Figure 8. The $P \cdot R$ dependence of $\alpha_{vv}$ values for the 2P (a) and 1N (b) systems. As expected (section 3.1) a change of slope takes place at a $P \cdot R \approx 0.3 \text{ hPa} \times \text{cm}$ corresponding to the average range of secondaries in the gas. While the simple model of section 3.1 (dotted line in the left panel) presents a saturation at large $P \cdot R$ values, the more precise Monte Carlo results (dots) indicate a small logarithmic growth.

number of secondaries that can be generated with very large energies (up to one half that of the primary electron) and are therefore able to escape even very large thicknesses.

5.2. Evaluation of the deposited energy

Whichever the procedure (experiment or calculation) used for the determination of the fluorescence yield, a precise evaluation of the energy deposited by the primary electron in the observation region is required. While the energy loss per unit path length for a primary electron...
Figure 9. Energy deposited by a primary electron per unit path length versus primary energy, for several values of $P \cdot R$ (continuous lines). In the limit of very large $P \cdot R$ values, deposited energy equals the energy loss of the primary electron predicted by the Bethe–Bloch theory (broken line). Density correction at atmospheric pressures was included.

can be accurately determined from the Bethe–Bloch law (above 1 keV), a significant fraction of that energy can escape the interaction region carried out by energetic secondary electrons or radiation. The determination of that ‘locally deposited’ energy, which depends on the geometry and size of the considered interaction region, can be carried out by means of Monte Carlo simulations.

The energy deposited by the primary electron per unit path length is related to the average energy deposited by secondary electrons $\langle E_{\text{dep}} \rangle$, obtained in the simulation, as

$$ (\frac{dE}{dx})_{\text{dep}} = N_{\text{air}} [ \langle E_{\text{dep}}^0 \rangle + \langle E_{\text{dep}} \rangle ] \sigma_{\text{ion}}(E). $$ (22)

Note that this equation is the same as (18) by replacing $\langle E_s \rangle$ by $\langle E_{\text{dep}} \rangle$. In figure 9, the results for our simple geometry are shown, i.e. a sphere of radius $R$. Deposited energy per unit path length versus primary energy for several $P \cdot R$ values is shown in the figure. As expected, the deposited energy for an unlimited medium, $P \cdot R \rightarrow \infty$, equals the energy loss predicted by the Bethe–Bloch theory.

The $P \cdot R$ dependence of the deposited energy is similar to that for the fluorescence emission described in section 5.1, that is, instead of a saturation at large $P \cdot R$ values, a smooth logarithmic growth is found. This behavior is also due to the very energetic secondary electrons with a large range and a significant probability of escaping the interaction region. While the number of those large energy secondaries is very small (decreasing as $1/E_s^2$ according to (16)), their integrated contribution to the energy balance is logarithmic and thus can be very significant at large primary energies even for very large $P \cdot R$ values.

5.3. The fluorescence yield

The ratio of the number of fluorescence photons to the deposited energy inside a given medium can be predicted from our simulations. The total number of fluorescence photons per unit path

Figure 10. (a) Fluorescence efficiency for the 2P(0, 0) band in the absence of collisional quenching against primary energy for several values of $P \cdot R$ (continuous lines). Dotted line represents the result for a very large medium. (b) The same for the 1N(0, 0) band.

The length emitted from a sphere of radius $R$ is given by (9) and (20). From these equations and (22), the following useful relation can be obtained for the fluorescence yield:

$$Y_{vv'} = Y_{vv}^0 \frac{1}{1 + P/P_v^0},$$  \hspace{1cm} (23)

where

$$Y_{vv}^0 = \frac{N}{N_{air}} \times \frac{(\sigma_{vv'}/\sigma_{ion}) + \alpha_{vv'}}{\langle E_{dep}^0 \rangle + \langle E_{dep} \rangle}.$$  \hspace{1cm} (24)

The values for $Y_{vv}^0$ have been computed for the 2P(0, 0) and 1N(0, 0) bands for a large range of primary energies and thicknesses $P \cdot R$. The variable $Y^0$ against $E$ for several values of $P \cdot R$ is shown in figure 10, whereas in figure 11 the dependence of $Y^0$ versus $P \cdot R$ is represented for various energies. For both figures, density correction at atmospheric pressure is included.

Several interesting features are shown in these figures. In the first place, for high energy (beyond the keV range) and for observation regions larger than about 1 hPa $\times$ cm, $Y^0$ is nearly constant. Nevertheless, a smooth dependence on primary energy is found. The value of $Y^0$ decreases with primary energy by about 10% in the range 1 keV–1 MeV and 4% in the interval 1 MeV–20 GeV for the 337 nm band (figure 10(a)). For the 391 band, the corresponding decrease is about 6% for the interval 1 keV–1 MeV and 1% for 1 MeV–20 GeV (figure 10(b)). On the other hand, a smooth growth of $Y^0$ with $P \cdot R$, smaller than 2% in the range 10–1000 hPa $\times$ cm, is also found for energies larger than 1 MeV (figure 11).

The above results support the assumption of proportionality between observed fluorescence and deposited energy for electrons with energy over a few keV. The contribution of the energy released by an extensive air shower by electrons with energy smaller than 1 MeV is only about 22% [53] and therefore the shown smooth dependence of fluorescence yield on electron energy has no relevant impact on the calibration of the fluorescence telescopes.

Within the above energy range, proportionality is predicted for observation regions over 10 hPa $\times$ cm, which is much thinner than the one used in any laboratory experiment. Below the
above limits (i.e. $E \lesssim 1$ keV, $P \cdot R \lesssim 10$ hPa × cm), fluorescence yield is strongly dependent on both the electron energy and geometry of the observation region.

Recently, several experiments have verified this proportionality under the usual laboratory conditions. To this end, two different techniques are used. The thick target experiment of the FLASH collaboration [19] compared the fluorescence light emitted in a shower induced by a high-energy electron beam on a thick target with the energy deposited at various depths. Proportionality is found within 5% uncertainty. On other hand, AIRFLY [20] has measured a constant fluorescence yield within several energy intervals (6–30 keV, 0.5–3 MeV, 3–15 MeV and 50–420 MeV) with uncertainties of 3–5%. Finally, MACFLY [11] measured a constant fluorescence yield at three energies (1.5 MeV, 20 GeV and 50 GeV) within about 13% uncertainty. It must be noted that those experimental results indicating a constant fluorescence yield are consistent with our results, since the predictions on slight energy dependence manifest in an energy interval much larger than the ones of these experiments.

In regard to the absolute values of the fluorescence yield obtained from our simulations, the results (at high energy and pressure) are $Y_{337}^0 = 350$ photons MeV$^{-1}$ and $Y_{391}^0 = 1600$ photons MeV$^{-1}$. As has already been discussed in section 3.2, while the dependence of the fluorescence yield on $E$ and $P \cdot R$ is accurately evaluated by our simulation, the calculation of the absolute values is subject to systematic uncertainties that are difficult to estimate.

6. Interpretation of laboratory measurements of air fluorescence

In this section, some experimental data will be analyzed taking into account the results presented in the previous section. Firstly, the pressure dependence of the effective optical cross sections would be useful for understanding some disagreements in the experimental determination of the $P'_v$ values measured by different techniques. Also, the pressure dependence and spatial distribution of air fluorescence at very low $P$ values can be properly interpreted with our model. Finally, our results will be used to make comparisons between measurements of air-fluorescence yield carried out under different conditions.

**Figure 11.** (a) Fluorescence efficiency for the $2P(0, 0)$ band in the absence of collisional quenching against the size of the observation region $P \cdot R$ for several primary energies. (b) The same for the $1N(0, 0)$ transition.
Figure 12. (a) Obtained \( P' \) values from a fit of the observed fluorescence in air to expressions (3) or (9) can differ significantly. (b) The resulting fit to observed \( \varepsilon_{vv}' \) values is usually similar in both cases, but the use of expressions (9) and (20) also justifies the absence of saturation at pressures much larger than \( P' \), due to the extra pressure dependence in \( \varepsilon_{vv}' \) from the \( \alpha_{vv}(P) \) factor.

6.1. Effect of secondary contributions on the determination of quenching parameters from fluorescence measurements

Neglecting the contribution of secondary electrons, the pressure dependence of the fluorescence yield can be described by the Stern–Volmer function (3). In this case, the fitting of the observed pressure dependence of fluorescence to this expression would allow the determination of \( P' \) parameters. Nevertheless, at near-atmospheric pressures, contributions from secondary electrons cannot be ignored and therefore the number of fluorescence photons generated per unit path length is given by (9). It must be noted that pressure dependence in that expression is not simple, as the \( \alpha_{vv}(E, P) \) factor included in \( \sigma_{vv}^{\text{eff}} \) (20) also depends on the geometry of experimental conditions.

Neglecting the \( P \) dependence of \( \sigma_{vv}^{\text{eff}} \) may lead to systematic errors in the measurement of characteristic pressures and this is illustrated in figure 12 using some \( \varepsilon_{vv}' \) experimental data in air from [10]. For both expressions (3) and (9), a plot of the product \( P\sigma_{vv}/\varepsilon_{vv}' \) versus \( P \) results in a straight line with \( 1/P' \) slope. Fits to both expressions are shown in figure 12(a). The pressure dependence of \( \sigma_{vv}^{\text{eff}} \) was obtained from (20) using the corresponding \( \alpha_{vv}(P) \) values calculated from our Monte Carlo simulation, assuming a size of the observation region of 2 cm. For comparison, figure 12(b) displays the usual plot of \( \varepsilon_{vv}' \) versus \( P \). While both fits in figure 12(b) are very close to each other, the slopes in figure 12(a) are clearly different. In fact, the resulting \( P' \) value obtained when taking into account the effect of secondaries decreases from 20.0 to 17.7 hPa. This smaller \( P' \) value is closer to that reported by Ave et al [13] (15.89 hPa), which uses the measured ratio of fluorescence intensities in nitrogen and dry air to cancel the effect of fluorescence from secondaries escaping the field of view.

Note that the pressure dependence predicted by (9) and (20) justifies the absence of saturation observed in plots of \( \varepsilon_{vv}' \) versus \( P \) (see e.g. [10]), due to the already discussed extra dependence of \( \varepsilon_{vv}'(P) \) on \( P \).
Figure 13. Taken from [17]. Fluorescence intensity as a function of gas pressure for 20, 100 and 250 eV energies of incident electrons. Symbols represent experimental data from [30]; curves are values from (11). A value of $R = 2$ cm was assumed for the only unknown parameter from the experiment (size of the interaction region). The curves were normalized to the same value at 0.1 Pa.

6.2. Nonlinear pressure dependency of fluorescence emission at low pressures

The above described quenching effects are not relevant at very low pressures $(P \ll P_{v})$ since the Stern–Volmer factor simplifies to unity. In those conditions, a linear pressure dependency of the observed fluorescence should be expected from expression (3). On the contrary, equation (9) indicates the presence of an extra pressure dependence even in the absence of quenching effects. This nonlinear dependency at very low pressures (a few Pa) was first experimentally observed by Fons et al [30], who suggested that it could be due to several interactions of primary electrons and/or to contributions from secondary ones. The contribution of secondary electrons as described above by equation (20) or (12) justifies quantitatively the experimental results. As an example, figure 13 presents some experimental data scanned from [30] for some incident electron energies. Our Monte Carlo simulation, assuming a value of 2 cm for the characteristic size of the interaction region, allowed us to easily justify the observed data. This check is also a validity test of our model. For more details, see [15, 17].

6.3. Spatial distribution of the emitted fluorescence

At very low pressures, the mean free path for high-energy electrons is very large and so experiments that involve the use of collimated electron beams observe only negligible beam dispersion when considering small interaction regions. If expression (3) were adequate for describing the excitation of gas molecules, all the fluorescence should be observed emitted from inside the electron beam. In contrast, fluorescence is also observed to come from regions well outside the path of the electron beam and, more significantly, emission depends on the observed wavelength.

According to the present interpretation of the processes involved, a significant amount of emitted fluorescence light should arise from secondary electrons generated in ionization processes. These are mostly low-energy electrons moving apart from the primary electron...
Figure 14. Spatial distribution of 391 and 337 nm fluorescence in the direction across a 30 keV electron beam at very low pressure (8 Pa). The overall beam size is indicated by a black arrow. Symbols represent the experimental data. Full lines are the results of the Monte Carlo simulation. For more details, see [15].

beam, and they justify the observed light outside the beam line. Taking into account the energy dependencies of $\sigma_{337}$ and $\sigma_{391}$ emission cross sections (figure 4(a)), one can also expect a significant amount of 337 nm photons arising from outside the primary beam, but only a few 391 nm ones ($\sigma_{391}$ is very small at low electron energies). In figure 14, this behavior is shown for 30 keV electrons at a pressure of 8 Pa in air. The Monte Carlo simulation carried out annotating the spatial distribution of emitted 337 and 391 nm photons, including the detailed geometry of the electron beam, justifies the experimental results (see [15] for more details of this experiment).

6.4. Comparison of experimental results

Several measurements of the absolute value of the fluorescence yield are available at present. As discussed in [16, 54, 55], the comparison between experimental data is not trivial since some authors have published results for a molecular band, while others have reported measurements in different wavelength intervals of interest for particular fluorescence experiments. In addition, sometimes this parameter is expressed in number of photons per electron and unit path length, whereas more recently, results have been given in photons per unit deposited energy (i.e. the fluorescence yield).

The analysis shown in the present work provides tools for a comparison among experimental results carried out at different conditions. In the first place, converting absolute $\varepsilon_{vv'}$ values to $Y_{vv'}$ fluorescence yields is not trivial because the relating factor $(dE/dx)_{dep}$ is not simply given by the Bethe–Bloch law, but must be obtained for each specific geometry, usually by Monte Carlo simulations. Nevertheless, as discussed in section 5.2, for near-atmospheric conditions, this geometrical dependence is very small and a rough estimate of the involved characteristic size (i.e. $R$) suffices. For instance, at atmospheric pressure, the difference in $(dE/dx)_{dep}$ between $R = 10$ cm and $R = 1$ m is below 5% for 1 MeV primary electrons. For that reason, the necessary values of deposited energy can be simply obtained from figure 9 using as a single parameter an estimated $P \cdot R$ value. In the case that $\varepsilon_{vv'}$ values had been
measured assuming that all the light is emitted from the primary track (i.e. neglecting the range of secondary electrons), a further correction on the optical efficiency to take into account the spatial distribution of fluorescence should be applied.

Comparison of the fluorescence yields for different wavelength intervals can be carried out as follows. Relative intensities \( I_{\nu'} \) within a band system are simply given by (7) and (8) in terms of the \( q_{X \rightarrow v} B_{\nu'} \) products and \( P_v' \) values. However, comparison of the observed fluorescence in wavelength intervals including both systems is dependent on the involved effective cross sections. Defining an effective cross section for the entire band system (1N or 2P),

\[
\sigma_{\nu\nu'}^{\text{eff}} = \sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} , \quad \Sigma_{\nu \nu'} q_{X \rightarrow v} B_{\nu \nu'} = 1 ,
\]

expression (9) can be rewritten as

\[
\varepsilon_{\nu \nu'}(P) = \frac{1}{kT} \sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} \frac{P}{1 + P / P_v'}
\]

or approximated by

\[
\varepsilon_{\nu \nu'}(P) \approx \frac{1}{kT} \sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} P_v'
\]

at large pressures \( P \gg P_v' \). Therefore, relative intensities are, in general, given by \( \sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} P_v' \).

Comparison of the fluorescence emission \( I_{\lambda} \) for a given band of wavelength \( \lambda \) with that from some desired wavelength interval \( I_{\Delta \lambda} \) can be then accomplished by means of the expression

\[
\frac{I_{\lambda}}{I_{\Delta \lambda}} = \frac{\sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} P_v'}{\Sigma_{\Delta \lambda} \sigma_{\text{sys}}^{\text{eff}} q_{X \rightarrow v} B_{\nu \nu'} P_v'}
\]

where \( P \gg P_v' \) has been assumed. For instance, the ratio between \( I_{337} \) and the total emission from 1N and 2P band systems would be

\[
\frac{I_{337}}{I_{1N} + I_{2P}} = \frac{\sigma_{2P}^{\text{eff}} q_{X \rightarrow 0} B_{00} P_0'}{\sigma_{2P}^{\text{eff}} \Sigma_{2P} q_{2P}^{\nu \nu'} P_v' + \sigma_{1N}^{\text{eff}} \Sigma_{1N} q_{1N}^{\nu \nu'} P_v'}
\]

where use has been made of the \( \Sigma_{\nu} B_{\nu \nu'} = 1 \) relation arising from the \( B_{\nu \nu'} \) definition (1).

While accurate values of \( q_{X \rightarrow v} B_{\nu \nu'} \) are available, significant disagreement exists between the \( P_v' \) results from different authors. For instance, \( P_{391}' \) values range from 1.3 hPa [32] to 2.9 hPa [13] or 5.0 hPa [10]. As a consequence, the evaluation of expression (28) or (29) using the \( \sigma_{1N}^{\text{eff}} \) and \( \sigma_{2P}^{\text{eff}} \) values obtained from our simulations combined with experimental \( P_v' \) values is unsafe. However, the \( \sigma_{\text{sys}}^{\text{eff}} P_v' \) factors resulting from experimental intensity ratios are subject to much smaller uncertainties. For instance, from the \( Y_{337} / Y_{391} = 3.6 \) ratio measured in [13] under near-atmospheric conditions, the value of \( \sigma_{1N}^{\text{eff}} P_{391}' / \sigma_{2P}^{\text{eff}} P_{337}' = 8.4 \) is inferred using (27). A very similar value, 8.0, results from the experimental \( Y_{337} / Y_{391} \) ratio observed by Nagano et al [10]. In contrast, the \( P_{391}' / P_{337}' \) ratios from both references are quite different (5.4 from [13] and 3.8 from [10]).

Other interesting ratios that result from (28) are \( I_{2P} / I_{1N} = 7.8 \) and \( I_{337} / (I_{1N} + I_{2P}) = 0.254 \). In addition, in table 1 the ratios \( I_{337} / I_{\Delta \lambda} \), obtained for the wavelength intervals used in several experiments are shown. The \( q_{X \rightarrow v} B_{\nu \nu'} \) products were taken from [23, 24] (using experimental intensity ratios from [13] gives very similar results as discussed in section 2.3 and figure 2).
Table 1. Comparison of the data on fluorescence yields. Experimental results are used to infer the value of the fluorescence yield for the 337 nm band at \( T = 293 \text{ K} \) and \( P = 1013 \text{ hPa} \) (the last column). See text and [55] for details.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>( \Delta \lambda ) (nm)</th>
<th>( T ) (K)</th>
<th>( P ) (hPa)</th>
<th>( \frac{I_{337}}{I_{\Delta \lambda}} ) (photons MeV(^{-1}))</th>
<th>( Y_{337} ) (MeV(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kakimoto et al [9]</td>
<td>300–400</td>
<td>288</td>
<td>1013</td>
<td>See text</td>
<td>0.278</td>
</tr>
<tr>
<td>Nagano et al [10]</td>
<td>337</td>
<td>293</td>
<td>1013</td>
<td>1.021 photons m(^{-1})</td>
<td>1</td>
</tr>
<tr>
<td>MACFLY [11]</td>
<td>290–440</td>
<td>296</td>
<td>1013</td>
<td>17.6 photons MeV(^{-1})</td>
<td>0.261</td>
</tr>
<tr>
<td>FLASH [12]</td>
<td>300–420</td>
<td>304</td>
<td>1013</td>
<td>20.8 photons MeV(^{-1})</td>
<td>0.276</td>
</tr>
</tbody>
</table>

Relative \( \sigma_{\text{sys}} \) values were obtained from the data in [13] (as mentioned above, using those from [10] would give very similar results).

The described procedures allow comparison of measurements under different conditions. As an illustration, several representative experimental data are presented and adapted for comparison in table 1. For this purpose, expressions (6) and (27) are used for converting the results at different temperatures to a reference one (e.g. 293 K). Fluorescence intensities in different wavelength intervals can be converted to that for the 337 nm band following the above described procedure. Expression (24) and data from figure 9 are employed for obtaining fluorescence yields after an estimate of the characteristic size of the interaction region. Finally, expression (23) converts the results at different pressures to a reference one (e.g. 1013 hPa).

In table 1, data from [11] and [12] are shown. For the comparison, only \( \Delta \lambda \), \( T \) and \( P \) corrections are required as both report fluorescence data in photons MeV\(^{-1}\). The value of \( \varepsilon_{337} = 1.021 \text{ photons m}^{-1} \) from [10] is also shown for comparison in the table. Moreover, the average value of the results from [9] corresponding to measurements of \( \varepsilon_{\Delta \lambda} \) in the 300–400 nm spectral interval at energies in the 1 MeV–1 GeV range is displayed in the table (see [55] for more details). For these measurements, characteristic sizes \( R \) of 5 and 10 cm for [10] and [9], respectively, have been assumed.

The resulting values in table 1 indicate general agreement in the \( Y_{337} \) with the differences being below 20%. Notice that the AIRFLY collaboration [14] has reported a preliminary result \( Y_{337} = 4.0 \text{ MeV}^{-1} \) (normalized at 293 K and 1013 hPa).

It is worth mentioning that in spite of the reservations indicated on the reliability of absolute values from our Monte Carlo calculations, the resulting \( Y_{337} \) value combined with the \( P'_{337} = 15.9 \text{ hPa} \) from [13] gives a fluorescence yield \( Y_{337} = 5.5 \text{ MeV}^{-1} \), which very favorably compares with the experimental ones in table 1.

7. Conclusions

The processes leading to fluorescence emission and energy deposition in air have been studied. The role of secondary electrons has been analyzed in depth. A simple model for the contribution of fluorescence from secondary electrons is able to account for many features of the atmospheric fluorescence. In particular, the 2P emission, dominant at high pressures, has been shown to be due to low-energy secondary electrons. In addition, a detailed simulation of the involved microscopic processes has been developed. An important ingredient of this simulation is the energy spectrum of secondary electrons, which has been well described by a simple analytical formula justified in section 4.
The simulation calculates both the deposited energy and the emitted fluorescence inside any geometry and thus the fluorescence yield. The results have been shown to be weakly dependent on the fine details of the geometry. Reasonable accuracy can be achieved assuming the interaction region to be a sphere of a given radial size. The precision of the absolute values of theoretically predicted fluorescence yields is limited by the uncertainties in the assumed molecular parameters. Nevertheless, our theoretical predictions are in reasonable agreement with available experimental data.

Also our simulation predicts that, under the usual experimental conditions, fluorescence intensity is nearly proportional to deposited energy. Nevertheless, for the 2P system, a smooth decrease of the fluorescence yield of about 10% in the range 1 keV–1 MeV and 4% in the interval 1 MeV–10 GeV is found. Taking into account that the contribution of the energy released by an extensive air shower by electrons with energy smaller than 1 MeV is only about 22% [53], the above smooth dependence has no impact on the calibration of fluorescence telescopes.

Apart from quenching, in a laboratory experiment, a further pressure dependence of the observed fluorescence results from secondary electrons escaping the field of view. As shown in the present work, \( P' \) values obtained from the measure of the pressure dependence of fluorescence intensity neglecting this effect are affected by systematic errors.

As expected, experimental relative intensities within a band system are proportional to \( q_{X \rightarrow v} B_{vv'} P'_{v'} \). Comparison of the fluorescence intensity in different wavelength intervals including both systems requires knowledge of the corresponding emission cross sections.

Values of \( \varepsilon_{\Delta \lambda} \) (photons per meter) can be converted to fluorescence yields (photons MeV\(^{-1}\)) by making use of our simulation results on deposited energy. In addition, a procedure has been shown to compare fluorescence intensities in different wavelength intervals from a combination of molecular parameters and experimental data. Following this procedure the results of several experiments have been compared.

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