

Corrigendum to

“Lightning-produced NO₂ observed by two ground-based UV-visible spectrometers at Vanscoy, Saskatchewan in August 2004” published in Atmos. Chem. Phys., 7, 1683–1692, 2007

A. Fraser¹, F. Goutail², C. A. McLinden³, S. M. L. Melo^{1,4}, K. Strong¹, and M. Van Roozendaal⁵

¹Department of Physics, University of Toronto, Toronto, Ontario, Canada

²Service d'Aéronomie du Centre Nationale de la Recherche Scientifique, Verrières le Buisson, France

³Environment Canada, Downsview, Ontario, Canada

⁴Space Science, Canadian Space Agency, Saint-Hubert, Québec, Canada

⁵Institut d'Aéronomie Spatiale de Belgique (IASB-BIRA), Brussels, Belgium

We have discovered two errors in our published paper “Lightning-produced NO₂ observed by two ground-based UV-visible spectrometers at Vanscoy, Saskatchewan in August 2004” (Fraser et al., 2007). The derivation of the VCD (vertical column density) of NO₂ contains an erroneous assumption, which is corrected in this corrigendum. In addition, the use of O₄ air mass factors (AMFs) instead of ozone AMFs gives a better calculation of the enhanced AMF due to path-enhancement by clouds associated with the thunderstorm. These corrections reduce the NO₂ flash production amounts calculated in the original paper by a factor of approximately seven, putting the estimates in line with the lower end of the range suggested by Schumann and Huntrieser (2007).

In the original manuscript, the lightning-produced VCD is calculated from the following two equations:

$$\text{DSCD}(\text{SZA}) = \text{VCD} \times \text{AMF}(\text{SZA}) - \text{RCD} \quad (1)$$

$$\text{DSCD}'(\text{SZA}) = \text{VCD} \times \text{AMF}'(\text{SZA}) - \text{RCD} \quad (2)$$

DSCD is the differential slant column density, RCD is the reference column density, and the primes indicate path-enhanced quantities due to the presence of clouds. Calculating the VCD from these two equations (giving Eq. (6) in the original paper) makes the erroneous assumption that the VCD does not change during the storm, which is precisely the increase that we are trying to calculate. This assumption is initially made to calculate the portion of the total NO₂

column that is due to path-enhancement through the clouds associated with the thunderstorm (and not due to lightning production). The equations that should be used to calculate the lightning-produced VCD are Eq. (2) and:

$$\text{DSCD}_{\text{obs}} = \text{VCD}_{\text{obs}} \times \text{AMF}_{\text{obs}} - \text{RCD}, \quad (3)$$

where the subscript obs indicates the observed DSCD and VCD. The AMF_{obs} will be changed by both the presence of clouds and the change in the NO₂ profile due to lightning-produced NO₂. The increase in the tropospheric NO₂ column is expected to be on the order of 10% (Winterrath et al., 1999): such an increase in the profile causes a 0–10% change in the total AMF as calculated by a radiative transfer model (McLinden et al., 2002). The cloud causes a minimum doubling of the AMF, and so the observed AMF can be approximated by the enhanced AMF' derived in Sect. 5.2 of the original paper.

Equations (2) and (3) can be expanded to consider the contributions from the stratosphere and the troposphere. Since the path and lightning-produced NO₂ enhancements are confined to the troposphere, the stratospheric VCD and AMF are unchanged:

$$\text{DSCD}' = \text{VCD}_{\text{strat}} \times \text{AMF}_{\text{strat}} + \text{VCD}_{\text{trop}} \times \text{AMF}'_{\text{trop}} - \text{RCD} \quad (4)$$

$$\text{DSCD}_{\text{obs}} = \text{VCD}_{\text{strat}} \times \text{AMF}_{\text{strat}} + \text{VCD}_{\text{obs,trop}} \times \text{AMF}'_{\text{trop}} - \text{RCD}. \quad (5)$$

The subscripts strat and trop refer to the tropospheric and stratospheric contributions, respectively. Solving these two



Correspondence to: A. Fraser
(amery@atmosp.physics.utoronto.ca)

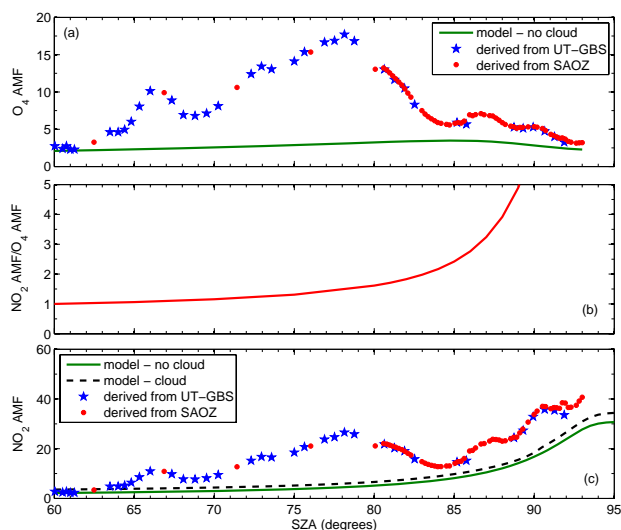


Fig. 1. (a) O_4 air mass factors from SCIATRAN and derived from the measurements. (b) Ratio of NO_2 -to- O_4 AMFs calculated using the radiative transfer models. (c) Same as (a), but for NO_2 . Also shown is the AMF calculated for the case of a cloud of $OD=70$ from 1–10 km using a radiative transfer model (McLinden et al., 2002).

equations for the change in the NO_2 VCD due to the production by lightning yields:

$$\Delta VCD_{\text{lightning}} = VCD_{\text{obs,trop}} - VCD_{\text{trop}} = \frac{DSCD_{\text{obs}} - DSCD'}{AMF'_{\text{trop}}}. \quad (6)$$

The enhanced tropospheric AMF is required to solve Eq. (6) for the VCD of NO_2 produced by lightning. In the original paper, the enhanced AMF' was calculated from the ratio of NO_2 to ozone AMFs. Since these are both primarily stratospheric species, this is not the ideal AMF' to use in solving Eq. (6). If the ratio of NO_2 to O_4 AMFs is used to calculate the AMF' , the derived AMF' can be used in solving Eq. (6), since O_4 is primarily a tropospheric species. The O_4 AMF was calculated from the SCIATRAN radiative transfer model (Rozanov et al., 2005, and references therein). The derivation for method two (Sect. 5.2 in the original paper) is followed, using the O_4 DSCDs and AMF instead of those for ozone.

Figure 1 updates Fig. 8 from the original paper, and shows the O_4 and NO_2 AMFs derived following method two. The O_4 AMFs increase by a factor of six: from a maximum near three to a maximum of 18. This is consistent with the increase in O_4 DSCDs shown in Fig. 5 in the original paper. The ratio of NO_2 to O_4 AMFs is shown in Fig. 1b. Unlike ozone and NO_2 , O_4 is a primarily tropospheric species, and its AMF peaks at about 85° . When the Sun is lower in the sky, the NO_2 AMF continues to increase, while the O_4 AMF is decreasing, which leads to the rapid increase in the ratio of the two AMFs. This behaviour, caused by the different shapes in the profiles of O_4 and NO_2 , will lead to inaccura-

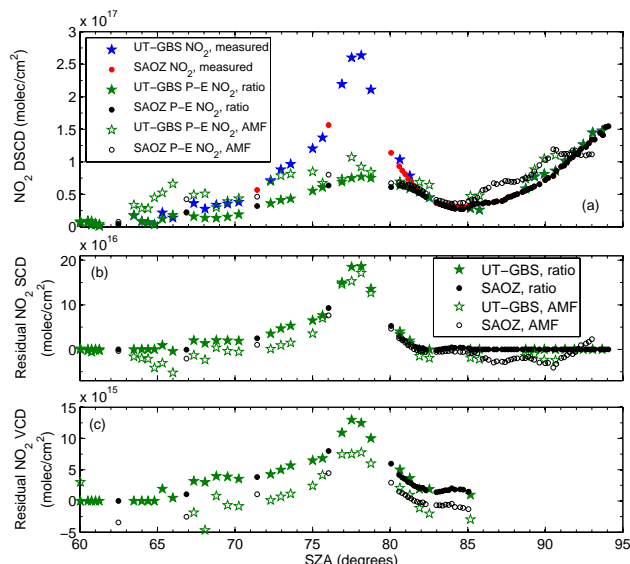


Fig. 2. (a) Measured total NO_2 DSCDs as well as the derived contribution from path-enhancement (P-E) for methods one (ratios to O_4) and two (derived AMFs). (b) Residual NO_2 SCDs attributed to lightning. (c) Lightning-produced NO_2 Δ VCDs calculated from the residual in (b) and Eq. (6).

cies in the NO_2 DSCD derived from this ratio, which is accounted for in the error assigned to the AMF ratio. Because of this rapid increase, the ratio method should not be used for SZAs larger than 85° . Figure 1c shows the NO_2 AMF' derived from the O_4 DSCDs, which is significantly larger than the NO_2 AMF' derived from ozone, shown in Fig. 8 of the original paper. This is likely a result of the increase in AMF being confined to the troposphere, something not considered in the original analysis.

Figure 2a updates Figs. 7 and 9 in the original paper, and shows the path-enhanced DSCD's derived using the two methods as well as the measured DSCDs from the two instruments (there is no change in the values derived using the ratio method). Prior to 67° , the value from the AMF method, which is an upper limit, exceeds the measured NO_2 DSCD, an indication that an assumption made in calculating the NO_2 AMFs is incorrect: most likely the ratio of the air mass factors is smaller than assumed. For all SZAs the DSCDs from the AMF method are larger than those from the ratio method, which is expected given the use of clear-sky ratios of AMFs.

Figure 2b shows the difference between the observed DSCDs and the calculated path-enhanced DSCD's, which is attributed here to production by lightning. Figure 2c shows the newly calculated Δ VCDs attributed to lightning using Eq. (6) and the newly derived NO_2 AMF' s. As expected from the larger AMF' s, the VCDs using the O_4 AMFs are smaller than those that appeared in the original paper. Integrating these curves yields the total amount of NO_2 produced

Table 1. Lightning-produced NO₂ columns (in 10¹⁷ molecules/cm²) and flash production amounts per cloud-to-ground (CG) flash (in 10²⁶ molecules/CG flash) calculated using Eq. (7) in the original paper.

Instrument	Excess NO ₂		Flash Production Amount	
	Ratio	AMF	Ratio	AMF
UT-GBS	0.98±0.18	0.40±0.07	1.06±0.34	0.43±0.14
SAOZ	0.77±0.14	0.40±0.07	0.83±0.27	0.43±0.14

by lightning, which is given along with the flash production amounts in Table 1.

These flash production amounts are universally smaller than the $(5.8\text{--}7)\times 10^{26}$ molecules NO₂/CG flash found by similar instruments (Franzblau and Popp, 1989; Langford et al., 2004). The best estimate of the production amount is found by using the ratio method, giving a range of $(0.83\text{--}1.06)\times 10^{26}$ molecules NO₂/CG flash. Taking into account the intra-cloud (IC) flashes reduces this range to $(0.13\text{--}0.17)\times 10^{26}$ molecules NO₂/flash. Schumann and Huntrieser (2007) compiled a list of 40 estimates of NO_x (NO+NO₂) production amount per flash (both CG and IC) found from theoretical calculations and ground, aircraft, and satellite-based observations. These values range between 0.04×10^{26} molecules NO_x/flash to 6.7×10^{26} molecules NO_x/flash. Based on this list, Schumann and Huntrieser (2007) conclude that the best estimate of the NO_x produced per flash is 1.5×10^{26} molecules NO_x/flash, with a range of $(0.2\text{--}4)\times 10^{26}$ molecules NO_x/flash. This result is not directly comparable with the NO₂ per flash amount derived in this work. However, the values derived here agree with the lower end of the range suggested by Schumann and Huntrieser (2007), while the amounts calculated by Franzblau and Popp (1989), Langford et al. (2004) and in the original paper (Fraser et al., 2007) are higher than this best estimate range.

References

- Franzblau, E. and Popp, C. J.: Nitrogen oxides produced from lightning, *J. Geophys. Res.*, 94, 11 089–11 104, 1989.
- Fraser, A., Goutail, F., McLinden, C. A., Melo, S. M. L., and Strong, K.: Lightning-produced NO₂ observed by two ground-based UV-visible spectrometers at Vanscoy, Saskatchewan in August 2004, *Atmos. Chem. Phys.*, 7, 1683–1692, 2007, <http://www.atmos-chem-phys.net/7/1683/2007/>.
- Langford, A. O., Portmann, R. W., Daniel, J. S., Miller, H. L., and Solomon, S.: Spectroscopic measurements of NO₂ in a Colorado thunderstorm: Determination of the mean production by cloud-to-ground lightning flashes, *J. Geophys. Res.*, 109, D11304, doi:10.1029/2003JD004158, 2004.
- McLinden, C. A., McConnell, J. C., Griffioen, E., and McElroy, C. T.: A vector radiative-transfer model for the Odin/OSIRIS project, *Can. J. Phys.*, 80, 375–393, 2002.
- Rozañov, A., Rozañov, V.-V., Buchwitz, M., Kokhanovsky, A., and Burrows, J. P.: SCIATRAN 2.0: A new radiative transfer model for geophysical applications in the 175–2400 nm spectral region, *Adv. Space Res.*, 36, 1015–1019, 2005.
- Schumann, U. and Huntrieser, H.: The global lightning-related nitrogen oxides source, *Atmos. Chem. Phys.*, 7, 3823–3907, 2007, <http://www.atmos-chem-phys.net/7/3823/2007/>.
- Winterrath, T., Koruso, T. P., Richter, A., and Burrows, J. P.: Enhanced O₃ and NO₂ in thunderstorm clouds: convection or production?, *Geophys. Res. Lett.*, 26, 1291–1294, 1999.