Supplemental Material: Derivation of simplified relationship for the Response (ρ)

Consider a box model of the NO_2 column, NO_2^T (x10^{15} molecules/cm^2) containing a point source of NO_x emissions, E^T (kTon/year). For simplicity, we use a uniform wind speed of *U* m/s and zero background of NO_2. We specify a chemical lifetime, τ_{chem}, for NO_2, and a conversion factor, *f*, of NO_x to NO_2 from the stack. Assuming that the box is in steady state, we can write the following mass balance:

$$E^T * f * S' = Flux_{out} + Sink \tag{S1}$$

where *S'* is a units conversion factor, *Sink* is the chemical sink in the box, and $Flux_{out}$ is the flux leaving the box of width, *w*. $Flux_{out}$ can be related to the average NO_2 column, assuming the point source is located in the middle and impacts half the cell, by:

$$Flux_{out} = 2\, w\, U\, NO_2^T * S'' \tag{S2}$$

where *S''* is a units conversion factor. The chemical sink in the box is the mass of NO_2 divided by the lifetime given by:

$$Sink = \frac{w^2 NO_2^T}{\tau_{chem}} * S'' \tag{S3}$$

Combining Equations S1-S3, we obtain:

$$E^T * f * \frac{S'}{S''} = \left(\frac{2U}{w} + \frac{1}{\tau_{chem}}\right) w^2\, NO_2^T \tag{S4}$$

The lifetime of NO_2 against wind transport, τ_{wind}, is given by:

$$\tau_{wind} = \frac{w}{2U} \tag{S5}$$

and the overall lifetime of NO_2 in the box is given by:

$$\frac{1}{\tau_{all}} = \frac{1}{\tau_{wind}} + \frac{1}{\tau_{chem}} \tag{S6}$$

Substituting Equations S5-S6 into S4, we obtain:

$$E^T * f * K = \left(\frac{1}{\tau_{wind}} + \frac{1}{\tau_{chem}}\right) w^2\, NO_2^T \tag{S7}$$

where K = S'/S''. Rearranging Equation S7, we get:

$$\rho = \frac{NO_2^T}{E^T} = \frac{K\, f\, \tau_{all}}{w^2} \tag{S8}$$

The larger contributor to τ_{all} is τ_{wind}, which is typically less than 1 hour for w ~ 20 km and U ~ 5-20 m/s, a typical range of wind speeds at plume height. Therefore, ρ is primarily dependent on U, which can be highly variable over time, and w. The dependence of ρ on chemistry is more complicated, as one needs to account for the nonlinear chemistry of NO_x and hydrogen oxide radicals ($HO_x = HO_2 + OH$) as discussed in Lamsal et al. (2011). Stavrakou et al. (2008) used NO_2 columns from GOME and SCIAMACHY and a CTM to infer a 3.8±1.8%/y decrease in NO_x emissions over the US from 1997 to 2006. They found that τ_{chem} increased by more than 15% over the northeastern US because of the emission reductions. Despite the complexity, ρ is relatively stable for each site so that ΔNO_2^T can be treated as linearly proportional to ΔE^T (e.g., Martin et al., 2003; Kaynak et al., 2009; Kim et al., 2009).

The difference between ρ as defined here and the parameter, β, presented in Lamsal et al. (2011) is that ρ establishes a linear relationship between the NO_2 columns and the stack emissions, and consequently has units related to those variables. In contrast, β establishes a relationship between $\Delta NO_2^T/NO_2^T$ and $\Delta E^T/E^T$, giving a dimensionless parameter. Our methodology is closer to that of Martin et al. (2003), who used a CTM to derive τ_{all} to relate the emissions to the column retrievals, but did not separate the lifetime into the components due to chemical transformation and wind transport.