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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations of HNO_3, ΣAN, ΣPN and NO_2 fluxes:evidence for rapid HO_x chemistry within a pine forest canopy" *by* D. K. Farmer and R. C. Cohen

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I entirely agree with referee #2 in the following, (a) Ďthe authors have to use questionable assumptions to reach a more or less consistent interpretation of the measured flux data", and (b) "a basic requirement of the used methodology is violated" (i.e., vertical constancy of fluxes and co-location of sources and sinks of the considered trace gases).

I agree also with the particular argument of both, referee #1 and referee #2, namely that the residence time used by the authors (400s) is much to long, the corresponding eddy diffusivity (0.34 ms-1) is much to low, and that the deduction of that values are

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based on a fairly (to my opinion: very) weak argument.

Application of eq. (10) (from Maartens et al.) - in that form how the authors make use of it - requires complete and homogeneous mixing of the layer from the forest floor (z=0) to the top of the canopy (z=hc) - which is never the case in which canopy ever. It is state-of-the-art knowledge in micrometeorology that values of the vertical distribution of in-canopy eddy diffusivities may range over 2-4 orders of magnitude. Therefore, estimating the aerodynamic resistance Rt (for eq. (10)) for the entire canopy from the inverse of HNO3 deposition velocity (which may be appropriate for the uppermost 1-2m of the canopy) is even not a zero-order approach - it is just wrong.

For their summertime data, the authors use a rather doubtful scaling argument for the required residence time, including winter/summertime friction velocity (ustar) and a very weak estimate of the height of the planetary boundary layer (an un-justified assumption of 1000m). I really wonder, why the authors have not used their own measured (micrometeorological) data to provide a sound and “chemistry independent” value of the required residence time (around canopy top, see argument(s) of referee #2).

The standard output of the used CSAT3 3-D sonic anemometer provides (and/or easily facilitates) data of friction velocity (ustar), variance of vertical wind speed (sigmaw), Monin-Obukhov length (L; from measured ustar, temperature and sensible heat flux), the height of the so-called zero-plane displacement (d), and the roughness length (z0). It is simple micrometeorological state-of-the-art knowledge to calculate the characteristic turbulent transport time (Tau, turb) over any given surface (canopy), which is an excellent scale for the corresponding residence time (see, Vila-Gerau Arellano & Dynkerke, 1992):

Tau,turb = kappa*[(z,meas - d) + z0] / (sigmawˆ2 * ustar)

Using kappa = 0.4 (v. Karman’s constant), I performed some back-on-the-envelope calculation of Tau,turb. Since the authors have even not provided one single value of any

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micrometeorological quantity (neither ustar or L, nor sigmaw), the necessary numbers of d, z0, L, and sigmaw have been estimated from the reported height of measurements (z,meas) and the height of canopy (hc) by the following (general) relations: d = 0.75*hc; z0 = 0.15*hc; sigmaw/ustar = 1.25*(1 - 3*z/L)ˆ(1/3) for L<0 (unstable conditions), and sigmaw/ustar = 1.25*(1+0.2*z/L) for L>0 (stable conditions), see Kirkman et al (2002). For any surface layer over any canopy there exist limiting relationships between ustar and z/L (e.g. Foken, 2003). Then Tau,turb could be calculated for any given realistic pair of ustar and z/L data. When varying z/L from -2 to +1, and ustar from 0.01 to 1 m/s (this roughly corresponds to horizontal wind speeds of 0.1 to 10 m/s) any calculated result for Tau,turb was definitely (much) less than 100s.

Anyway, I am sure, that the authors (when using their measured sonic anemometer data of ustar, L, sigmaw, etc.) will either confirm (or rebut) my estimates easily.

However, <100s residence time (summertime) would force the authors to require OH concentrations even (at least) 4 fold higher as those (already pretty high) concentrations which they are postulating.

Finally, another three comments:

(1) like referee#2, I consider the application of photostationary state calculations to get (not measured) NO concentrations from measured NO2 concentrations definitely as risky (if not wrong). It is widely accepted common sense among those groups which are performing simultaneous NO-NO2-O3 flux measurements (even outside of California) that close to (emitting and/or absorbing) surfaces the photostationary equilibrium is never fulfilled.

(2) since more than 15 years it is state-of-the-art knowledge in micrometeorology, that immediately over tall canopies there exists the so-called roughness layer (a sub-layer of the surface layer), where Monin-Obukhov similarity laws fail. In this layer, for a given flux, the corresponding (scalar) gradient is about half (or even less than half) of that which would be calculated over a smooth surface. Since the pioneering paper of

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Garrat (1978) there is ample evidence that the depth of the roughness layer may extend up to 2-3 times of canopy height (hc). Garrat (1992), Shuttleworth (1989), Simpson & Thurtell (1998), and Moelder et al (1999) have developed correction formulas for the semi-empirical M-O stability functions to consider the influence of the roughness layer. However, following the procedure of Moelder et al, the authors may develop their own (site specific) correction functions making use of simultaneous measured CO2 fluxes and gradients.

(3) application of the "modified Bowen ratio" method (while the authors call it "Bowen ratio" method, the term "Bowen ratio" is usually limited to the ratio of sensible and latent heat flux) deserves also a very careful check of the necessary assumptions. These particularly address the demanding requirement of the co-location of sources and sinks of the considered trace gases. Assuming a-priori co-location without any (plausible or real) proof makes the application of the modified Bowen ratio method at least doubtful. Furthermore, as attractive this method may be, its application in the roughness layer requires additionally, that the above mentioned correction functions for the M-O-stability functions can be regarded as identical between the individual trace gases (see Foken, 2003).

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 7087, 2007.