

Interactive comment on “Depletion of gaseous polycyclic aromatic hydrocarbons by a forest canopy” by S.-D. Choi et al.

S.-D. Choi et al.

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Re (1) While the chemical analysis ... taken into account.

The focus of the study presented in the manuscript is on finding evidence of the uptake of semi-volatile organic chemicals in a forest canopy. The flux calculation is one aspect of the data interpretation, but not the major one, and this is reflected in the detail provided in the method section.

During the experiment, we experienced various technical difficulties with an aging profile and data acquisition system (which was replaced later in 2003). Due to these circumstances the eddy diffusivities based on CO₂ and H₂O were rather noisy, and given that our confidence of the accuracy of the temperature profile is much higher, we chose to use this for our K calculations. Daytime maxima and diurnal patterns of

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KCO₂, KH₂O and KHeat agreed well, so other than introducing additional noise the use of KCO₂ or KH₂O would have produced the same results. We are aware of the fact that the sonic temperature represents a "speed of sound" based temperature which is functionally similar (but not quite equal!) to the virtual temperature:

$T(\text{sonic}) = T(1+0.51q)$ [Schotanus et al., Boundary Layer Met. 26, 81-93, 1983]

This correction to the heat flux (which is part of the standard operating procedure for Ameriflux and other flux networks) is typically a few percent or less. The crosswind correction is done automatically by the CSAT sonic. In our case the q correction was not applied to H (given a relatively noisy latent heat flux), but given its relatively small effect the outcome of our modeling exercise is not affected.

Re (2) Another problem related ... methodological factors.

As outlined in section 3.3., 24-hour integrated samples make it difficult to apply a real-time MBR approach, since it is highly unlikely that stationary conditions will be encountered throughout the 24 hours. On top of this, the aforementioned technical difficulties we had resulted in only 4 of the 12 days with PAH data having continuous eddy flux data. This is why we chose to run through a modeling exercise that would provide us with a fair order-of-magnitude estimate of the fluxes.

Re: The upper and lower quartiles ... precision of the results.

As we mentioned in section 2.4, despite of the large uncertainty associated with the use of median values of KHeat, we tried to provide a reasonable estimate of the range within which the real flux resides. We believe that the use of medians and upper and lower quartiles can fulfill our purpose. We could obviously calculate PAH fluxes using KHeat data covering more than 50% (e.g. 5-95% etc), but it would hardly yield more insight.

Re: I would like to see ... assumed diurnal cycle scenarios.

We could have written this manuscript without any calculations of fluxes and deposi-

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tion velocities. It would still be a worthwhile contribution by being the first study to observe vertical concentration gradients of PAHs in the atmosphere, and by presenting the first direct observational evidence of the uptake of gaseous PAHs by a forest canopy. However, we felt it is worthwhile to make full use of the data and attempt at least a semi-quantitative interpretation yielding estimates of important kinetic parameters. The purpose of this paper is not to provide accurate flux estimates; a different experiment design would have been necessary to do this. The manuscript is very forthright throughout about the very large uncertainty of the estimated fluxes and deposition velocities. It is stated clearly that the presented data are order of magnitude estimates. It is not clear to us what a systematic uncertainty analysis would yield other than the confirmation that the data should be regarded as order of magnitude estimates. Therefore, we chose not to dwell on the micrometeorological details because, again, this is not the thrust of the manuscript.

Re: (3) Deposition velocities ... are realistic.

This is a good suggestion. We calculated the bulk aerodynamic resistances (u_{33m}/u^2). The median mid-day limit to deposition velocities (assuming a zero surface resistance) is on the order of 10-15 cm s⁻¹. This does indeed suggest that the surface resistance for these PAHs is relatively small.

Re: (4) Even though Fuentes et al ... using MBRM.

These are more appropriate references:

Businger, J.A.: Evaluation of the accuracy with which dry deposition can be measured with current micrometeorological techniques. *Journal of Climate and Applied Meteorology* 25, 1100-1124, 1986.

Wesely, M.L., and Hicks, B. B.: A review of the current status of knowledge on dry deposition. *Atmos. Environ.* 34, 2261-2282, 2000.

Re: The issue of Simpson et al. ... the related uncertainties.

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According to the detailed study by Simpson et al. (1998), as well as those conducted at Borden by Fuentes et al. (1996) and Kaharabata et al. (1999), level B is certainly in the RSL, and even level A will still be affected. However, as the reviewer points out, the MBR method has the advantage that as long as the vertical distribution of the sources and/or sinks of temperature and the PAH is the same, the gradients for both temperature and the PAH concentrations are underestimated by the same extent and the effect cancels out. This does not account for the fact that eddy diffusivities for sensible heat and gas transfers have some fundamental physical differences (especially at non-neutral stratifications), but for practical purposes they are usually assumed to be the same [cf. Stull, R.B. 1988. An introduction to boundary layer meteorology. Reidel Publishing Co., Dordrecht, 666 pp.]

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 2359, 2008.

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