**Atmospheric
Chemistry
and Physics
Discussions**

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Comment

Interactive comment on "Depletion of gaseous polycyclic aromatic hydrocarbons by a forestcanopy" *by* S.-D. Choi et al.

S.-D. Choi et al.

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Re: The factor of 5 reduction in SVOC ... observed in the field.

We do not entirely agree with MacLeod in this regard. Clearly, the calculated relative importance of advection and deposition depends on the size of the model domain. In a very small model domain, advection dominates no matter how fast competing processes of deposition (or degradation) may be. However, the time constant for deposition remains the same with increasing size of the model domain, as long as surface cover and the height of the atmosphere is assumed to remain the same. That implies that with a 10-fold increase in the size of the model domain, the deposition loss also increases by an order of magnitude. On the other hand, advective losses diminish in relative importance with increasing model size. If we assume our model domain to be circular, advection losses only increase by a factor of 3.16, i.e. the square root of

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10, for every order of magnitude increase in the size of the model region. In other words, deposition losses are increasing in importance relative to advection losses with increasing model size.

It is correct that the calculation in Wania and McLachlan (2000) assumed that the area upwind of the modeled region is also forested, and thus ignored the effect of advective dilution (essentially assuming an infinitely large forested area). We therefore agree that the factor of 5 reported in Wania and McLachlan (2000) represents the maximum possible effect of a forest cover on air concentrations of persistent organic chemicals. However, we continue to believe that this effect can be much larger than 1.2 in areas of significant forest coverage (i.e. considerably larger than the 50,000 km2 forest used by MacLeod (2003), i.e. continental scale forests such as the boreal forests of Siberia and Canada. This was indeed confirmed in global scale model calculations by Su and Wania (2004).

Re: Scenario I is ... during the day.

Reviewer MacLeod refers to what is called the "quasi-laminar resistance (qlr)" - the resistance of a thin layer on the surface on a scale where molecular processes become important. This layer may be intermittent, depending on the level of disturbance by the turbulent layer next to it. However, our gradient measurements are looking at two points outside this quasi-laminar layer. We cannot say much about the gradient across this layer, except that it will be smaller during the day - possibly correlated with a larger gradient between levels A and B to compensate. Given the fact that the deposition velocities calculated from the PAH gradients with the MBR method are close to the inverse of the aerodynamic resistance (equivalent to the maximum eddy "conductance"), surface resistances (including the qlr) are probably secondary for gaseous PAH.

Re: Considering all of the uncertainties ... scenario in Table 2).

We agree with reviewer MacLeod. In fact, this sentiment has already been expressed (with those very words!) in the manuscript: "the flux estimate can only be expected

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to deliver an order of magnitude estimate". We don't see what additional information could be derived from a more formal quantification of uncertainty. For example, the use of multiple scenarios for the diurnal variability of dC/dz was meant to convey the uncertainty that is introduced by the poor temporal resolution of the air concentration measurements. We feel that the fluxes and deposition velocities presented in the paper are clearly identified as order of magnitude estimates (for example:"provide a rough estimate of a range within which the true flux is likely located").

Re: One interesting thought ... Su et al.

Attempting such a back-of-the-envelope budget approach, let's assume a 10 m thick layer of active canopy, which we cut off from the outside world through hypothetical lids above and below. With a deposition velocity of 10 cm/s, species x in this slab is depleted on a time scale of 100 s (e-folding time). With a deposition velocity of 1 cm/s, it takes 1000 s. For typical aerosols between 0.1 and 1 um diameter (accumulation mode), deposition velocities have been found to be between 0.1 and 1 cm/s, so it might take up to 10,000 s [cf. Petroff, A., Mailliat, A., Amielh, M., Anselmet, F., 2008. Aerosol dry deposition on vegetative canopies. Part I: Review of present knowledge. Atmos. Environ., in press, doi:10.1016/j.atmosenv.2007.09.043]. According to the Lagrangian footprint model by Strong et al. (2004) the typical air parcel residence time in the canopy at Borden is on the order of 0.5 to 1 minute in the upper part of the canopy. So the reviewer MacLeod's intuition is right on: gaseous PAH is depleted on the same time scale as the mixing, whereas particle deposition is a factor of 10-100 slower.

Re: Figure 2: The three measurements ... these data?

The 100 % particle-bound fraction during those three days is a result of the levels in the gas phase being below the limit of detection, while very small concentrations (< 5 pg/m3) in the particle phase could be deteced. Note, that the concentration of anthracene are generally much smaller than those of other compounds, and therefore closer to the limit of detection. (see Table S4 in the Supporting Information for detailed

data).

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

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