

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

### **Anonymous Referee #1**

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#### General evaluation

This paper presents an interesting experimental study of the dry deposition of PAH compounds to a Canadian deciduous forest in spring. Depletion of gaseous PAHs by the forest canopy is analyzed based on daily samples of vertical PAH gradients and the micrometeorological "modified Bowen ratio" technique. The results provide direct evidence of the uptake of semi-volatile organic compounds by vegetation. The removal rates were quantified as mass flux densities and deposition velocities for three compounds. Deposition velocity is an important parameter, needed for estimating the atmospheric residence times and surface removal rates, but previous data on PAHs are very limited. Thus the results of this paper constitute a valuable dataset.

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The topic of the study is very suitable for the scope of ACP. The paper is clearly organized and written. The supplement makes it possible to present a large amount of detailed supporting material, while keeping the main text clear and concise. However, there are some issues, mainly related to micrometeorology, that need further clarification and revision. I would recommend publishing this paper in ACP, if the following questions can be addressed in an adequate manner.

### Major comments

(1) While the chemical analysis is described in sufficient detail, the same is not true for the meteorological measurements. Because of the rather cursory description of these data, it is not possible for the reader to evaluate the derivation of the turbulent exchange coefficient (eddy diffusivity). The specification of instrumentation and the eddy covariance flux calculation principles are missing. In particular, it would be important to explain how the temperature gradient was measured.

Furthermore, it should be explained in more detail why the eddy diffusivity of sensible heat, rather than water vapour or carbon dioxide, was chosen ("heat flux was best behaved", p. 2370, is much too vague), given the fact that the vertical temperature gradients are often very small.

It should also be noted that, in principle, the "sonic" temperature provided by a sonic anemometer is not equal to air temperature but approximates the virtual temperature (corresponding to buoyancy rather than sensible heat flux); it is not clear whether this difference has been taken into account.

(2) Another problem related to eddy diffusivity is the averaging procedure. It is not explained why hourly medians calculated over a 51-day period were used instead of the actual eddy diffusivities of each PAH sampling day. As Figure S7 indicates, there is considerable variation in the hourly values, due to both meteorological and methodological factors.

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The upper and lower quartiles were used for representing an uncertainty range, but these results are not commented in any way. Furthermore, the quartiles only cover 50% of the data and thus provide an optimistic estimate of the precision of the results.

I would like to see a more detailed and systematic uncertainty analysis of the results. This should combine the uncertainties related to both the eddy diffusivity and concentration gradients including the assumed diurnal cycle scenarios.

(3) Deposition velocities exceeding 10 cm/s seem very large, especially as diurnal averages. The upper limit of possible deposition velocities can be estimated by assuming that there is no surface resistance to uptake, that is, by assuming that the deposition velocity is only controlled by the aerodynamic resistance. As the authors have all the required data available, a comparison of the derived deposition velocities with the calculated (inverse of) aerodynamic resistances could be used as an additional quality assurance procedure for demonstrating that the large deposition velocities are realistic.

(4) Even though Fuentes et al. (1996) and Simpson et al. (1998), cited on p. 2366, do present an overview of the theoretical background of flux-gradient relationships, neither of these papers presents a discussion of the modified Bowen ratio method (MBRM) used in the present paper. The method used by Fuentes et al. (1996) and Simpson et al. (1998) depends on the universal flux-gradient functions, which do not need to be specified when using MBRM.

The issue of Simpson et al. (1998) is the roughness sublayer (RSL; also discussed by Fuentes et al.), which limits the applicability of the Monin-Obukhov similarity (MOS) theory near the top of a forest canopy. While MBRM is not limited by RSL effects to the same extent as the methods relying on the universal MOS functions, scalar similarity (identical eddy diffusivities) must be assumed. While this may be appropriate above RSL, the situation is more complicated within RSL, due to the heterogeneity of the source/sink distributions and differences in these distributions between different scalars. Therefore it would seem appropriate to mention that at least the lower

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measurement level for gradients is located within RSL and to discuss the related uncertainties.

#### Minor comments

p.2361, lines 7-9: It is unclear if this is only due to depletion by dry deposition; please rephrase.

p.2362, section 2.1: Data on the development of the leaf area index during the measurement period would be useful.

p.2362, lines 16-17: "when PAH uptake in the forest canopy is expected to be largest"; please explain why.

p.2364, line 24: Replace "diffusion" by "turbulent".

p.2365, lines 12-13: Kurt-Karakus et al. (2006) is not a suitable reference for justifying the assumption that  $KC = K_{Heat}$  (especially so as they assume  $KC = K_m$ , the eddy diffusivity of momentum).

p.2366, line 13: Please explain the summation symbol.

p.2366, line 15: Please explain what is meant by "high winter values".

p.2366, line 21: Please present the correlation coefficient.

p.2367, lines 27-29: Are these two sentences basically saying the same thing? And shouldn't one expect significant gradients due to dry deposition to active vegetation?

p.2368, lines 1-2: It is unclear how large gradients were considered "strong", as compared to those required for determining the eddy diffusivity.

p.2371, lines 7-9: I am unable to understand this explanation. If PAHs are revolatilized from the forest canopy during the daytime, then this would result in a negative  $dC/dz$ , not a larger positive  $dC/dz$ .

p.2372, Eq. 4: As downward flux is negative by definition, a minus sign is missing from

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the right-hand side.

p.2372, lines 16-17: The results differ from the previous data by a factor of 1.5-4. Is this really a "reasonable agreement"?

p.2376, line 4: Replace "31" by "31(SI)" to indicate the special issue.

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

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