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Interactive comment on “Mercury dynamics in the Rocky Mountain, Colorado, Snowpack” by X. Fain et al.

Anonymous Referee #2

Received and published: 30 December 2012

Dear Fain et al.

This manuscript presents a field study on GEM fluxes in alpine surface snow. The data allowed comparison GEM gradients to those of ozone, nitrogen oxides and carbon dioxide. The results indicate the importance of snow chemistry both in the dark and photolytically driven on gas-phase mercury concentrations in non-polar snow-packs. These are very important findings. The topic is highly relevant to a large audience. The conclusions are sound and the data are carefully discussed. I recommend publication after tackling some minor issues:

Page 15425 line 4: For some remote places, ocean currents seem to play an important role as source of Hg (Fisher, J. A., Jacob, D. J., Soerensen, A. L., Amos, H. M., Steffen, A. and Sunderland, E. M.: Riverine source of Arctic Ocean mercury inferred

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from atmospheric observations, *Nature Geosci.* 5(7), 499–504, doi:10.1038/ngeo1478, 2012.)

Page 15247 line 9: Could you give more details on the actual snow cover and snow-fall events during your study periods?

Page 15432 line 20: Does the "entire measurement campaign" refer to the 2009 data (as implied by the next sentence) or to 2009 and 2011-2012. If so, why are data from 2011-2012 not included in Figure 2?

Page 15433 line 8 and page 15435 line 15: How deep is the photolytic zone? Could you estimate this based on structural characterisation of the snow-pack. I'd guess this is less than 60 cm which would further support your conclusion in the importance of transport processes.

Page 15435 lines 26: I'm surprised that Hg(II) complexes with Cl⁻ can be photolyzed by solar light, as the absorption is well below 300 nm; rather around 200 nm. (Kunkely, H., Horvath, O. and Vogler, A.: Photophysics and photochemistry of mercury complexes, *Coordination Chemistry Reviews*, 159 IS -, 85–93, doi:doi: DOI: 10.1016/S0010-8545(96)01307-0, 1997.) Are there newer data to support this mechanism?

Page 15436 line 20. This numerical simulation was done for diffusion through porous snow taking porosity and tortuosity into account, wasn't it?

Page 15438 line 2 You might consider adding this reference: Douglas, T. A., Sturm, M., Simpson, W. R., Blum, J. D., Alvarez-Aviles, L., Keeler, G. J., Perovich, D. K., Biswas, A. and Johnson, K.: Influence of Snow and Ice Crystal Formation and Accumulation on Mercury Deposition to the Arctic, *Environ. Sci. Technol.*, 42(5), 1542–1551, doi:10.1021/es070502d, 2011.

Page 15439 line 4 I was wondering, if the snow is permeable throughout the whole measurement campaign or if ice layers might have formed that inhibit diffusive transport. Do CO₂ or NO data support the permeability?

BGD

9, C6941–C6943, 2012

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Page 15439 line 10 Please specify “adsorption energy”. If you refer to DHads this would not be sufficient to estimate the partitioning, as it neglects the entropy. You would need to discuss DGads or K. Also, both quantities are standard quantities. Please give the standard state. (Donaldson, D. J., Ammann, M., Bartels-Rausch, T. and Pöschl, U.: Standard States and Thermochemical Kinetics in Heterogeneous Atmospheric Chemistry, *J. Phys. Chem. A*, 120418133855003, doi:10.1021/jp212015g, 2012.)

Page 15443 As temperatures approach 0°C I would suggest that liquid water might be present. Impurities and geometric constraints in grain boundaries can stabilize water well below 0°C. The role of the quasi-liquid layer is much more speculative (Bartels-Rausch, T., Jacobi, H. W., Kahan, T. F., Thomas, J. L., Thomson, E. S., Abbatt, J. P. D., Ammann, M., Blackford, J. R., Bluhm, H., Boxe, C. S., Domine, F., et al.: Relationship between snow microstructure and physical and chemical processes, *Atmos. Chem. Phys. Discuss.*, 12(11), 30409–30541, doi:10.5194/acpd-12-30409-2012, 2012.)

Page 15443 line 21. Your data set give the unique opportunity to show the importance of freeze-induced reactions in the field. Just for curiosity, have you looked for episode where your snow-pack might undergo freezing (at temperatures below 0°C due to melting point depression). Do those data indicate a decrease in GEM?

Interactive comment on Biogeosciences Discuss., 9, 15423, 2012.

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