

Interactive comment on “Vertical advection and nocturnal deposition of ozone over a boreal pine forest” by Ü. Rannik et al.

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This strikes me as an important polemic, both particularly relevant to the ozone deposition issue at hand, and also for which a forum such as ACPD is well-suited to hashing out a resolution.

As the editor points out the Seinfeld and Pandis text - clearly an essential reference for atmospheric chemists - exactly defines concentration as a synonym for density (mass/volume). Conversely however, fundamental textbooks for atmospheric physicists exactly define concentration as a dimensionless proportion of molecules (e.g., Batchelor, *An Introduction to Fluid Dynamics*, 2000). Climate scientists too, dating at least to Keeling’s famous 1960 publication regarding CO₂ concentrations, have used the term to express a dimensionless proportion. Given long-standing traditions for

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these competing meanings for "concentration" in different atmospheric disciplines, it seems unlikely that any one group of atmospheric scientists will impose their definition on others. Pointedly however, in the context of diffusive transport relevant to the (eddy covariance) manuscript at hand, it is the Seinfeld and Pandis text that uses the term falsely.

It may appear to be a matter of general agreement that Fick's law relates the diffusive molecular flux to a gradient in concentration. However, thorough examinations of fluid transport phenomena (see Batchelor but also Bird, Stewart, and Lightfoot, Transport Phenomena, 2002) specify Fickian diffusion as a function of gradients in molecular proportions, i.e. the dimensionless definition of concentration. The version of Fick's law presented in the Seinfeld and Pandis text is erroneous, precisely because the authors have used a definition for concentration that is incongruent with the physical process being described. The difference can be quite significant for vertical transport in the atmosphere, since profound vertical gradients in density (the chemists' concentration) exist for every gas species due to gravity and fluid compressibility, but often have nothing to do with diffusion.

Concerning diffusion of reactive gases and specifically for the case of ozone deposition, whatever the definition it is incorrect to specify "concentration" universally as (a) the variable whose fluctuations and gradients define diffusive transport phenomena, and (b) the determinant of gas-phase reaction rates. I reiterate therefore that concentration is an ambiguous term whose continued use, if truly necessary, should be accompanied by a careful expression of units wherever possible. Far preferable would be for chemists to use the unambiguous term density and for physicists to refer to the molar fraction, rather than continuing to use the same word while referring to different scalar quantities.

Finally, the fact that the fluid constituent proportion termed the mixing ratio has different meanings for physical meteorologists (who specify it in mass terms, appropriate to modeling the outcome of mixing) and atmospheric chemists (for whom it would seem to be synonymous to the molar fraction) is also potential source of misunderstanding.

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In the interest of clarity, this would seem to require some resolution for the disciplines of Atmospheric Chemistry and Physics. In this case however, the difference is merely a scaling factor according to molecular masses, and at least has no effect on the fundamental meaning of physical laws.

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