

In regards to the approach the authors use to describe transport, it should be pointed out that their equation 3 can be rearranged to be made up of a purely diffusive term and one advection term, and this is what is numerically being solved (simplifying to one direction, letting $f_{w,e}=1$):

$$\frac{\Delta C}{\Delta t} = \frac{F_A(\Delta x U_{pos})}{\Delta x^2} \left[\frac{C_{i-1} - 2C_i + C_{i+1}}{\Delta x^2} \right] + F_A(U_{pos} - U_{neg}) \left(\frac{C_i - C_{i+1}}{\Delta x} \right)$$

The first term is a purely diffusive term with a diffusivity of $F_A(\Delta x U_{pos})$, which shows that it is dependent on the grid size, which is not likely appropriate here for a few reasons, including: 1. (It is likely more proper to use the average of the positive and negative components, instead of just U_{pos} . This leads to the second term being a centered difference, potentially adding more diffusion.) Diffusion should not be dependent upon the grid size, and 2. This means that the effective transport distance, by diffusion, is also grid size dependent, 3. Effectively (discussed more below), exposures in a city are mainly determined by an effective horizontal diffusion, not net transport due to advection. In terms of the dependence on grid size, material can be transported twice as far (or fast) just because of the choice of grid size doubling. The second term is also important in that it shows that the equation is asymptotically correct (e.g., the case where U is only positive or negative) only if $F_A=1$ (they have set it to 2), and is likely not mass conservative without being set to 1. For the current solution approach, the actual equation they solve more closely resembles:

$$\frac{\partial c}{\partial t} + 2\nabla u_{net}c = \frac{\partial}{\partial x}(2U\Delta x) \frac{\partial c}{\partial x} + \frac{\partial}{\partial y}(2V\Delta y) \frac{\partial c}{\partial y} + \frac{\partial}{\partial z}(K_z) \frac{\partial c}{\partial z} + R + E - D$$

The effective horizontal diffusion is likely about 300,000 m²/s ($U \sim 5$ m/s; $dx \sim 30,000$ m). This makes the effective Pe # (UL/D) about 0.002 (highly diffusion dominated) for transport in a city of size 100 km. In essence, they now take that the difference between the average positive and negative velocities to become a stochastic fluctuating quantity, and that the magnitude and direction is independent and uncorrelated to the other velocity components. It would be instructive to compare their solution with a few analytical solutions for simplified one dimensional transport cases (e.g., where the wind blows west at 5 m/s for 5 months and then east at 4 m/s for 7 months, with a source in the center, and chemical depletion of 0.1 per day), and maybe a two dimensional case as well ($U_{pos}=5$, $U_{neg}=2$; $V_{pos}=5$, $V_{neg}=4$, each half the time; source in the center).

Thus, at this point, the authors need to address the issues of the very large numerical diffusivity and the lack of asymptotic agreement with the governing equation, before this approach should be utilized.