RE: Impact of modified turbulent diffusion of PM2.5 aerosol in WRF-Chem simulations in Eastern China
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General Comments - Peter Taylor

The authors have changed the title, as suggested, and made a series of changes but I still have concerns about some missing details. There are also sections, including the abstract, where some careful language edits could clarify the text.

The Richardson number dependent eddy diffusivity (Eq2, 5) for particles in stable conditions (*Ri* > 0) was developed and discussed in Jia et al (2021b). In that paper Ri is said to be a **gradient** Richardson number "where *f(Ri)* is empirical stability function of gradient Richardson number (*Ri*)." but **is not defined**. The definition given in the present paper (Eq 1) is a bulk Richardson number based on differences relative to "surface level values". For winds are these 0 or $U_{10}$? Tracking down Ri use by different authors is always a problem. Esau and Byrkjedal (2007) use finite difference approximations to the local gradient Ri, and discuss issues associated with their accuracy, " Another important aspect of the vertical resolution is a numerical approximation of strongly-curved vertical profiles on coarse meshes using finite-difference numerical schemes." The current paper seems to use Esau and Byrkjedal's TDC functions for heat and momentum, but with a bulk *Ri*.

Fig 3 of Jia et al (2021b) shows the data used to develop Eq 5. There is a lot of scatter, near zero values of $f_c$ for several data points with *Ri* < 0.2. For *Ri* > 0.2 there are some large (~0.6) experimental $f_c$ values. Bottom line is that I would have very little confidence in the $f_c$ formulation proposed by Jia et al (2021b).

If implementation in WRF-CHEM as a tuning exercise produced well documented, convincing results that could certainly be of interest but I am not convinced by the material here. We are given information on the impacts of changes to the TDC for particles on model results. They do appear to improve overall comparisons with observed average values but I would like to see much more detailed discussion. One set of daily PBL height values (Fig 6) are given as a time series but it would be informative to see more sample time series data, of $PM_{2.5}$ concentrations and fluxes at observation points, and with hourly resolution to see day-night differences. Start with some time series comparisons and then worry about the overall statistics. It is hard to take output from a large, complex meteorological model like WRF-CHEM in order to see the impact of tuning one of the internal equations but we should be shown more of the details.

The confusion about exactly what is used as *Ri* is concerning. Does the model *Ri* correspond to the value used from the measurement data from Jia et al (2021b)?

Many of my concerns relate to that previous GRL paper but it would not be good for ACP to further encourage the use of a dubious result.

I am also concerned about the sources of $PM_{2.5}$. We are simply told to believe their inventory. Are we looking at $PM_{2.5}$ from smoke stacks and subsequent chemical transformation, or is some of it road and other dust? Is the ground surface always a sink or can it be a source? How can we be sure that the differences between model prediction and observations are not related to the source inventory?

Detailed comments

line 105 Were these PM2.5 flux measurements compared with model predictions?

line 114 Good to calculate observed vertical fluxes, but are the results used or compared with the modelled fluxes. Is the ground a source or sink of $PM_{2.5}$ in these simulations?

line 123 Note that Eq(1) is a bulk Ri, and explain what are the surface level values, $u_s$, $v_s$, $\vartheta_{vs}$.

line 141 .. horizontal **grid** resolution.... and the factor 5 is rather larger than usually applied.

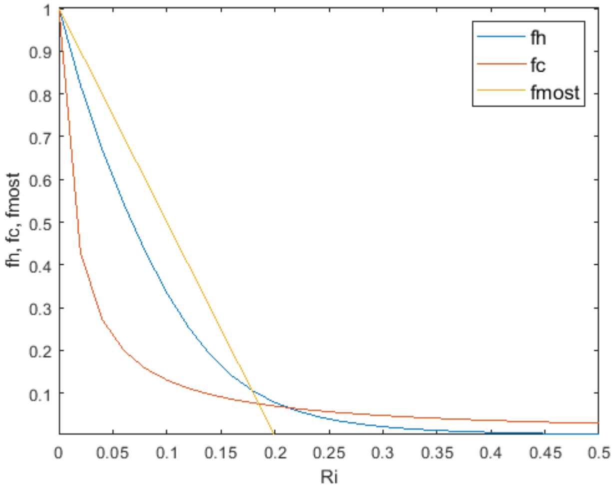
line 147 ? **Single** layer UCM , and explain UCM. If measurement sites are urban then this could be an important issue.

line 165 A 64 h spin up seems long for WRF (typically 12 h for meteorology) but may be needed for WRF-CHEM if initial concentrations are unknown, also why 91? 64 + 24 = 88? Can you explain reasons for the long spin up time. Are results sensitive to this?

line 175 Is the 0.01 minimum value common to the original and new schemes?

line 176 If one wants u = 0 at z =0 then one can set $l = k(z+z_0)/(1 + kz/\lambda)$. where $z_0$ is a roughness length. There is no mention of roughness length until line 353, but it can be an important parameter and should be different for momentum, heat and $PM_{2.5}$. It is present in WRF and should be discussed. It could be linked to the 0.01 minimum in Eq (2)?

line 186 It is worth comparing the current $f_h$ and $f_c$, with the expression one would get using MOST, if we use the standard form $1/f_h = \varphi_h = 1 + 5\ z/L$, and relationship with Ri from Garratt (p52), and assuming L = 0 for Ri > 0.2. A quick plot is below with $f_{most} = 1/\varphi_h$.



For moderate values (<0.2) of *Ri*, MOST and $f_h$ are both significantly larger than $f_c$ so that the modified TDC is significantly reduced relative to MOST or earlier assumptions with $K_c = K_h$.

For Ri > 0.25, $f_c > f_h$ as noted on line 188, but how often does this occur - in reality and in the model. In the model there may be confusion between gradient and bulk Ri. Can we see a pdf of *Ri* values?

line 199 "avoids the inapplicability of MOST". This "inapplicability" should be explained, maybe it is because we are not necessarily in a constant flux layer? MOST need not require $f_c = f_h$.

line 208 How is "night" defined in forming these averages?

line 223 Why the big difference in 2014? I am puzzled by why "relative bias" and "absolute bias" % differences are different. I assume that these are night-time values? Based on hourly data?

From Fig 3 the relative and absolute bias values seem to be of opposite sign at some locations, A detailed definition of these quantities should be provided. I can guess but a few equations would help.

line 235 So n in Fig 4 is 31*24? I am not familiar with Taylor diagrams. Is the vertical axis from the model and the horizontal the observations?

line 261 Can you say anything about deposition to the surface, which could be a critical removal process, or a source?

line 287 There is a lot to be said about keeping models as simple as possible. What exactly do you have in mind as a " turbulence-aerosol two-way feedback module". But not needed here.

line 344 This seems to be the first mention of "source" and we are expected to accept that " there is no way to use other more elaborate inventories to quantify the uncertainty caused by emissions".

line 395 Maybe tell us what the mean absolute errors in hourly or daily $PM_{2.5}$ values (μg $m^{-3}$) are in order to see how significant these bias improvements are.

line 398 " Therefore, the pollutant concentration is reduced near the surface and better mixed in the whole layer, increasing the pollutant concentration in the upper level." Are there any upper level measurement to validate this effect?

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Reference.

Garratt, J.R., (1992) The atmospheric boundary layer, Cambridge, UK