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Comment on acp-2021-435

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Referee comment on "Impact of modified turbulent diffusion of PM_{2.5} aerosol in WRF-Chem simulations in eastern China" by Wenxing Jia and Xiaoye Zhang, Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-435-RC2>, 2021

This is the latest in a series of papers on air quality and aerosol issues in China that these authors have been involved with. There are considerable similarities between this manuscript and material in the cited GRL paper, Jia et al (2021b). The basic idea is that turbulent diffusion of heat differs from diffusion of momentum, of other scalars, and of aerosol particles. This is not a new idea and is generally dealt with in terms of ϕ functions of z/L , where L is the Obukhov length ($-u_{\text{ref}}^3/[k(g/\theta)\langle w'\theta' \rangle]$). Here u_{ref} is the friction velocity, k is the Karman constant, θ is potential temperature and $\langle \dots \rangle$ denotes a time or ensemble average. In the current paper Eq (1), for eddy diffusivities (TDC), includes a stability function $f(\text{Ri})$ which differs between heat, f_h , momentum, f_m and particles, f_c . This could be analogous to $\phi_M(z/L)$, $\phi_H(z/L)$ differences in the Monin-Obukhov approach.

Eq (1) also includes a constant, 0.01, without any explanation or specification of units. It also appears to be absent in Jia et al (2021b). Given that the mixing length expression used in Eq (1) does not include a roughness length, z_0 , then one interpretation could be that $0.01 = ku_{\text{ref}}z_0$. The issue is then whether there should be different roughness lengths for momentum, heat and aerosol.

The present paper, and Jia et al (2021b) only present $K(\text{Ri})$ relationships for $\text{Ri} > 0$ (stably stratified conditions, while the WRF-CHEM model is run for day and night situations. Although the focus is on night-time conditions, we need to know what is done when $\text{Ri} < 0$ ($\partial\theta/\partial z < 0$). Is $f_c = f_h$ in those cases?

The authors claim (line 134) that Monin-Obukhov similarity theory (MOST) is inapplicable and later (line 150) that "If the MOST is applicable, it indicates the turbulent mechanisms of heat, water vapor and particles are the same,..." without substantiating that erroneous claim. MOST is based on the idea of a surface boundary-layer with fluxes of heat and momentum being approximately independent of height. It is widely used within the surface layers of models such as WRF and ECMWF models. Dimensionless velocity and temperature gradient functions, $\phi_M(z/L)$, $\phi_H(z/L)$, based on MOST (e.g. Garratt, 1992, Eq

3.33 a,b) can differ and counter the line 150 claim. Admittedly these are in the unstable, $Ri < 0$, $L < 0$ case but there is nothing inherent in MOST to say that they should be equal in stable conditions.

Negative remarks about MOST, here and in Jia et al (2021b) are used to support diffusion models based on gradient Richardson number, Ri (without ever defining it). The problem with diffusion coefficients based on $Ri [= (g/\theta)\partial\theta/\partial z/[(\partial U/\partial z)^2 + (\partial V/\partial z)^2]]$ is that velocity and temperature gradients have strong z variation, basically proportional to $1/(z + z_{0q})$, where z_{0q} is the roughness length appropriate to the quantity involved, close to the surface and finite difference calculations of gradients can be very unreliable. Meanwhile L is constant in a constant flux layer. In deeper layers, the flux Richardson number ($Rf = (g/\theta)\langle w'\theta' \rangle / (\langle u'w' \rangle \partial U/\partial z + \langle v'w' \rangle \partial V/\partial z)$) is widely used. For aerosol in surface layers, MOST and Buckingham's Pi theorem, could allow an additional dimensionless variable w_s/u_{*0} , where w_s is the gravitational settling velocity, and could lead to interesting results allowing for variation between quantities being diffused by turbulence. Many models account for this via a deposition velocity for aerosol which combines the effects of turbulent diffusion and gravitational settling. The formulations of Zhang et al (2001) are a good example. Farmer et al (2021) show that deposition velocities, for micron sized particles, can vary significantly with particle diameter, underlying surface and friction velocity, and that "our understanding ... is poor".

An addition relative to Jia et al (2021a) are some data on correlation coefficients (Fig 2). It was not clear exactly what these data were averages of but from Ren et al (2020) we can find some details, which should be provided here. We should be told at what height these flux measurements are from. On average R_{wt} has a strong diurnal cycle while R_{wc} has a mean close to 0 implying minimal vertical flux. I assume that $R_{wc} > 0$ implies an upward flux of aerosol. Since much of the discussion is in terms of $PM_{2.5}$ "pollution" and (line 95) gives information on anthropogenic emissions I had been thinking in industrial emission terms rather than land surface dust as the major component of the aerosol. Some clarification on this would be helpful.

Winter 2013-2017 Eastern China runs with the modified diffusion formulation for stable stratification are also new. We are told that $PM_{2.5}$ concentration predictions are reduced. We are not really told why or where the $PM_{2.5}$ particles go? Is the dust source reduced? Does more $PM_{2.5}$ deposit on the ground, mix higher in the boundary layer or spread more widely in the horizontal? We are told nothing about deposition velocities but my guess would be that they average to zero (some + and some -) since Fig 2c shows near zero R_{wc} values.

Overall this is a scientifically weak paper. It is not well written and has a strange title. That being said it is on an appropriate topic for ACP, it has some new results, relative to Jia et al, 2021b, although the basic idea and much of the discussion is similar. With Major Revision, less background material and fewer unnecessary references, plus the addition of some missing details, on $Ri < 0$, $PM_{2.5}$ sources and sinks, surface boundary conditions, plus modelled aerosol budgets, then it could be publishable.

As I see it, a more appropriate title could be "Impact of modified turbulent diffusion of PM2.5 aerosol in WRF-Chem simulations in Eastern China". I cannot see that the manuscript demonstrates that a "Unified treatment of scalars is a missing source of turbulent diffusion on PM2.5 concentration in WRF-Chem"

References

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