Response to Referee #1

Question1:

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*3/[k(g/θ)<w'θ'>]. Here u* is the friction velocity, k is the Karman constant, θ is potential temperature and <...> denotes a time or ensemble average. In the current paper Eq (1), for eddy diffusivities (TDC), includes a stability function f(Ri) which differs between heat, fh, momentum, fm and particles, fc. This could be analogous to φM(z/L), φH(z/L) differences in the Monin-Obukhov approach.

Response1:

We feel great thanks for your professional review work on our article. As you are concerned, there are several problems that need to be addressed. It is true that the turbulent diffusion of momentum, heat and particles are different, and this is not a new idea. Previous studies have to study turbulent diffusion of particles by Computational fluid dynamics (CFD) model (Derudi et al., 2014; Fiates et al., 2016; Longo et al., 2019), experiment (Altunbas et al., 2002; Flesch et al., 2002; Sofiev et al., 2009), Reynolds-averaged Navier-Stokes equation (RANS) approach (Sini et al., 1996; Gualtieri, et al., 2017) and other means. However, for the mesoscale model, especially for a two-way coupled atmospheric-chemistry mesoscale model (e.g., WRF-Chem and GRAPES_CUACE), few people pay special attention to the turbulent diffusion of particles. Just as what you said, the stability functions (i.e., f_h, f_m, f_c) is analogous to $\varphi(z/L)$ (i.e., $\varphi_m(z/L)$ and $\varphi_h(z/L)$). Nonetheless, the turbulent diffusion of particles in the current mesoscale model is expressed by turbulent diffusion of heat, which has some errors. Therefore, based on this idea, we first establish the turbulent diffusion relationship of particles based on Mixing length theory by using observational data, and then apply it to the mesoscale model (Jia et al., 2021). In our last article, we focused on establishing the turbulent diffusion relationship of particles through the observation data, and then added it into the model, which was preliminarily verified only by the results of 2016. In this manuscript, we mainly analyze the turbulent diffusion of particles from the perspective of model. The long-term simulation results are used to verify the reliability of the previous results, and the existing uncertainties are analyzed to provide the basis for future work.

Question2:

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, z0, then one interpretation could be that 0.01 = ku*z0. The issue is then whether there should be different roughness lengths for momentum, heat and aerosol.

Response2:

Actually, 0.01 refers to the minimum value of turbulent diffusion coefficient (TDC) in the model. For detailed parameter setting, refer to lines 653-841 (i.e., Subroutine EDDYX) of Program (i.e., module_bl_acm.F in WRF-Chem v3.9.1). Here, we have taken partial screenshot for reference (Fig. R1), where EDYZ0=0.01.

```fortran
  SUBROUTINE EDDYX(DTPBL, ZF,  ZA,     MOL, PBL,  UST,                 &
                   US,    VS,  TT,  THETAV, DENSX, PSTAR,              &
                   QVS,   QCS, QIS, DSIGFI, G, RD, CPAIR,              &
                   EDDYZ, EDDYZM, its,ite, kts,kte,ims,ime,kms,kme )

!**********************************************************************
!   Two methods for computing Kz:
!   1.  Boundary scaling similar to Holtslag and Boville (1993)
!   2.  Local Kz computed as function of local Richardson # and vertical
!       wind shear, similar to LIU & CARROLL (1996)
!
!**********************************************************************
!
!-- DTPBL           time step of the minor loop for the land-surface/pbl model
!-- ZF              height of full sigma level
!-- ZA              height of half sigma level
!-- MOL             Monin-Obukhov length in 1D form
!-- PBL             PBL height in 1D form
!-- UST             friction velocity U* in 1D form (m/s)
!-- US              U wind
!-- VS              V wind
!-- TT              temperature
!-- THETAV          potential virtual temperature
!-- DENSX           dry air density (kg/m^3)
!-- PSTAR           P*=Psfc-Ptop
!-- QVS             water vapor mixing ratio (Kg/Kg)
!-- QCS             cloud mixing ratio (Kg/Kg)
!-- QIS             ice mixing ratio (Kg/Kg)
!-- DSIGFI          inverse of sigma layer delta
!-- G               gravity
!-- RD              gas constant for dry air (j/kg/k)
!-- CPAIR           specific heat of moist air (M^2 S^-2 K^-1)
!-- EDDYZ           eddy diffusivity for heat KZ
!-- EDDYZM          eddy diffusivity for momentum KM
!----------------------------------------------------------------------
!----------------------------------------------------------------------

      IMPLICIT NONE

!.......Arguments

!... Integer
      INTEGER,  INTENT(IN)   ::    its,ite, kts,kte,ims,ime,kms,kme
!... Real
      REAL , DIMENSION( ims:ime ),           INTENT(IN)  :: PBL, UST
      REAL ,                                 INTENT(IN)  :: DTPBL, G, RD
      REAL , DIMENSION( kts:kte ),           INTENT(IN)  :: DSIGFI
      REAL , DIMENSION( its:ite ),           INTENT(IN)  :: MOL, PSTAR, CPAIR

      REAL , DIMENSION( ims:ime, kms:kme ), INTENT(IN)  :: US,VS, TT,   &
                                                           QVS, QCS, QIS, DENSX
      REAL, DIMENSION( its:ite, kts:kte ), INTENT(IN) :: ZA, THETAV
      REAL, DIMENSION( its:ite, 0:kte )  , INTENT(IN) :: ZF

      REAL , DIMENSION( its:ite, kts:kte ), INTENT(OUT) :: EDDYZ,EDDYZM

!.......Local variables

!... Integer
      INTEGER  :: ILX, KL, KLM, K, I

!... Real
      REAL     :: ZOVL, PHIH, WT, ZSOL, ZFUNC, DZF, SS, GOTH, EDYZ
      REAL     :: RI, QMEAN, TMEAN, XLV, ALPH, CHI, ZK, SQL, DENSF, KZO
      REAL     :: FH, FM
      REAL     :: WM, EDYZM, PHIM
!... Parameters
      REAL, PARAMETER :: RV     = 461.5
      REAL, PARAMETER :: RC     = 0.25
      REAL, PARAMETER :: RLAM   = 80.0
      REAL, PARAMETER :: GAMH   = 16.0 !Dyer74 !15.0  !  Holtslag and Boville (1993)
      REAL, PARAMETER :: GAMM   = 16.0 !Dyer74
      REAL, PARAMETER :: BETAH  = 5.0   !  Holtslag and Boville (1993)  BETAM = BETAH
      REAL, PARAMETER :: KARMAN = 0.4
      REAL, PARAMETER :: P      = 2.0   ! ZFUNC exponent
      REAL, PARAMETER :: EDYZ0  = 0.01  ! New Min Kz
      REAL, PARAMETER :: PR     = 0.8   ! Prandtl #
```

Figure R1. Partial screenshot of program (i.e., module_bl_acm.F in WRF-Chem v3.9.1).

Question3:

The present paper, and Jia et al (2021b) only present K(Ri) relationships for 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 Ri < 0 ($\partial\theta/\partial z < 0$). Is fc = fh in those cases?

Response3:

We are very sorry that we did not clearly explain the situation under all conditions. We have described the calculation principle of turbulent diffusion of particles under stable and unstable conditions, and see section 2.3 for details.

Question4:

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, φM(z/L), φH(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.

Response4:

After the reviewer's suggestions, we have deleted the content of this section (i.e., temperature-particles transport dissimilarity). The connection between this section and other contents in the text is not very good, which is a little abrupt here. According to your suggestions later, we have also modified the title of the article, and the contents of the article is more in line with your suggested title.

Question5:

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/θ)∂θ/∂z/[(∂U/∂z)2 + (∂V/∂z)2] is that velocity and temperature gradients have strong z variation, basically proportional to 1/(z + z0q), where z0q 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 ws/u^*, where ws 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".

Response5:

We are sorry that some concepts have not been clearly stated, and we have revised them. In fact, we have not made negative comments on MOST. In addition, previous studies have shown that the inapplicability of the MOST in the stable boundary layer (Edwards et al., 2020; Grachev et al., 2012), and our method is to avoid using the MOST under stable conditions. Moreover, we also evaluated the uncertainty difference between the two methods in the previous paper (Jia et al., 2021). We mentioned in our previous article that the TDC calculated by MOST and PBL height under stable conditions is uncertain, so we use the Mixing length theory to replace it. While under the unstable conditions, we still use MOST to calculate the TDC. Therefore, MOST and Mixing length theory are used in the model at the same time. We quite agree with you on the effects of gravitational settling, as Zhang et al. (2001) said, the parameterization of particle dry deposition is also extremely important. With increasing particle size, particle inertia and gravity cannot be neglected, but these inertia and gravity effects are neglected for particles smaller than 10 μm in diameter (Fratini et al., 2007). Therefore, we do not consider the gravity effect of particles here, but we added discussions on gravitational settling. In the future, we will use long-term simulation results to verify the difference of aerosol process decomposition in detail.

Question6:

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 Rwt has a strong diurnal cycle while Rwc has a mean close to 0 implying minimal vertical flux. I assume that Rwc > 0 implies an upward flux of aerosol. Since much of the discussion is in terms of PM2.5 "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.

Response6:

We have deleted this section. We hope we can have a separate article to study the turbulent transport between momentum, heat and particles in more detail based on the observational data (this work is in progress).

Question7:

Winter 2013-2017 Eastern China runs with the modified diffusion formulation for stable stratification are also new. We are told that PM2.5 concentration predictions are reduced. We are not really told why or where the PM2.5 particles go? Is the dust source reduced? Does more PM2.5 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 Rwc values.

Response7:

In fact, we have explained in Figure 5 that the pollutant concentration was reduced in the surface layer, and it was mixed to the upper level, and the pollutant concentration increases in the upper level. Firstly, there is no change in emission sources, so the impact of emission sources can be excluded. Secondly, the pollutant concentration decreases near the surface layer, so more pollutants do not deposit on the ground. At the same time, the pollutant concentration increases in the upper level, and it is mixed in the boundary layer. Finally, if the pollutants are transported in the horizontal direction, when the underestimation of pollutant concentration at a certain station is improved, there will be no unified change driven by the winds in the Eastern China. Therefore, the pollutants are better mixed in the boundary layer.

Question8:

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, PM2.5 sources and sinks, surface boundary conditions, plus modelled aerosol budgets, then it could be publishable.

Response8:

Compared with the previous article (Jia et al., 2021), some of the results may be a little similar, mainly because we use the long-term simulation to verify the previous result, which are consistent with the previous results. In comparison, the previous article pays more attention to establish the turbulent diffusion relationship of particles based on the observational data. While this study pays more attention to the uncertainty analysis of the model results. According to your nice suggestions, we have made extensive corrections to our manuscript.

Question9:

As I see it, a mote 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".

Response9:

We have revised the title according to your suggestion.

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