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:

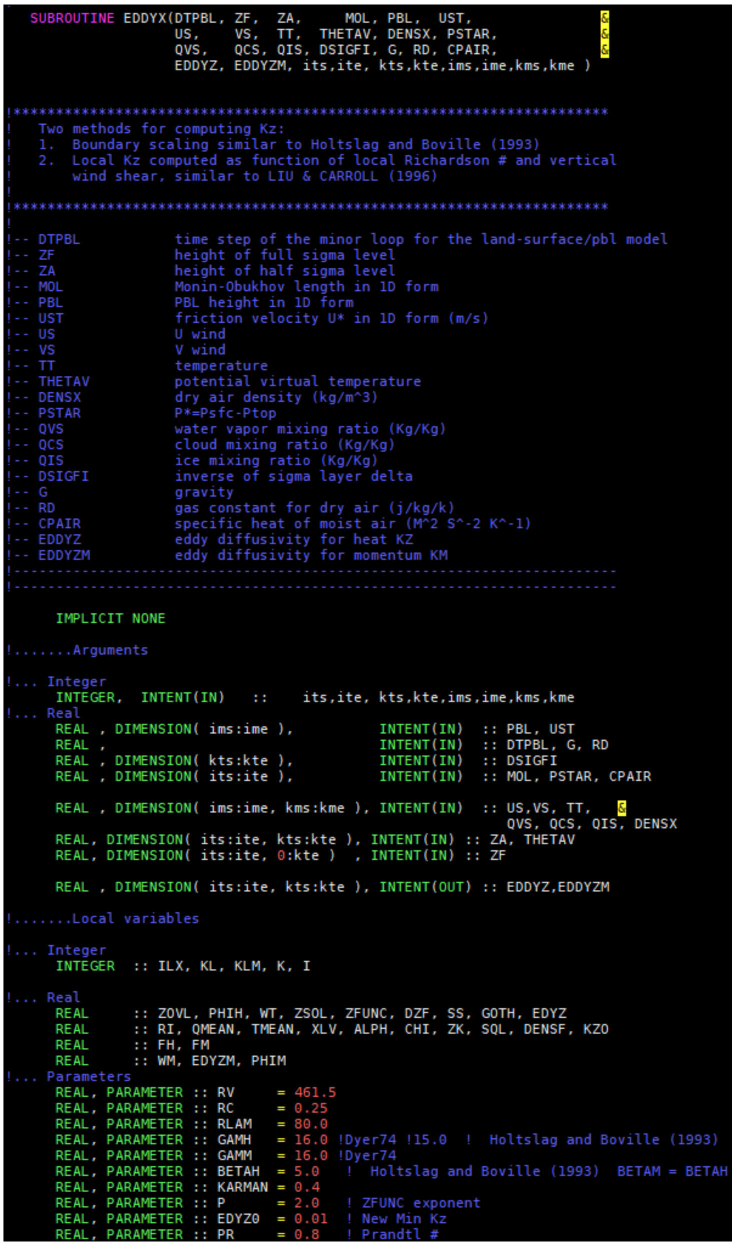
We feel great thanks for your professional review work on our article. As you are concerned, there are several problems that need to be addressed. It is true that the turbulent diffusion of momentum, heat and particles are different, and this is not a new idea. Previous studies have to study turbulent 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.



```
   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 (∂θ/∂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, $\varphi M(z/L)$, $\varphi H(z/L)$, 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.

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 $[= (g/\theta)\partial\theta/\partial z/[(\partial U/\partial z)2 + (\partial V/\partial z)2]$ is that velocity and temperature gradients have strong z variation, basically proportional to $1/(z + z0q)$, 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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Response to Referee #2

General Comments:

The scheme of turbulent mixing process of particles in the atmospheric boundary layer directly influences the particle concentration predicted by the numerical models. In this paper, the authors introduce a new scheme of turbulent diffusion coefficient, which is different from that for scalars, to describe the turbulent mixing process of particles in WRF-Chem model. The new scheme is only for stable conditions, while under unstable conditions the scheme is not changed. The results show that the new scheme can improve the prediction of particle mass concentration when compared to the output of model using the original scheme. However, the physics behind the results seems problematic. The numerical simulations show that the model using the new scheme reduces the overestimated PM2.5 concentration simulated using the original scheme. But the new scheme has a smaller turbulent diffusion coefficient (TDC) than the original one. It means that the model using the new scheme should predict larger PM2.5 concentration than using the original scheme. I cannot understand why a smaller TDC will lead to smaller PM2.5 concentration in the stable atmospheric boundary layer. So I think the authors should make additional analysis on their simulation results and tell us why a smaller TDC can result in smaller PM2.5 concentration under stable conditions (I mean, the authors should tell us the real reason for the reduced PM2.5 concentration simulated using the new scheme). Additionally, in this paper the authors provide some evidence to argue that the new scheme is reasonable for describing the turbulent mixing process of particles. I think the evidence is not convincing. In Section 3 the analyses of observational data show that the behaviors of transport efficiencies for heat and particle are different. But this result does not support the use of a smaller TDC for turbulent mixing of particle. As for the discussions in Section 5, the provided evidence does not tell us the physics that the model using the new scheme can have the better performance in predicting the PM2.5 concentration under stable conditions. So I think the presentation of this paper is not well done and the conclusions are unconvincing. My recommendation is major revision.

Response:

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. According to your nice suggestions, we have made extensive corrections to our pervious draft, and the detailed corrections are listed below. Actually, we want to make a point, which is also the most important point, that is, the turbulent diffusion coefficient (TDC) in the new scheme is larger than that in the original scheme, not smaller. Therefore, using the new TDC will lead to lower pollutant concentration in the stable boundary layer.

Specific Comments:

1) One of my major concerns is why the model using a smaller TDC can predict smaller PM2.5 concentration under stable conditions. Eqs. (2)–(5) give the new scheme of eddy diffusivities (the authors call eddy diffusivity as TDC) for heat, momentum and particle under stable conditions, in which the eddy diffusivity for particle K_c is different to that for heat K_h. Actually, the difference between K_c and K_h is embodied by the different Ri-dependant functions, f_c and f_h, as expressed in Eq. (5) and Eq. (3) respectively. The two equations indicate that in the new scheme f_c is smaller than f_h (in the original scheme f_c is equal to f_h, as expressed in Eq. (3)). Therefore the new scheme has smaller eddy diffusivity for particle than the original scheme. To our knowledge, the smaller eddy diffusivity means the weaker turbulent transport, which results in higher concentration of air pollutant when other conditions are the same. However, the simulations in this paper show that the model using the new scheme predicts smaller PM2.5 concentration than the model using the original scheme. My question is why. I think it is the most important issue in this paper. The authors should do additional analyses on the simulation results to find the reasons and tell us what the reasons are.

Response:

Here, we would like to point out that the turbulent diffusion coefficient (TDC) of the new scheme is greater than that of the original scheme, which can dispel your doubts. We agree with you that the difference between K_c and K_h is embodied by the different

Ri-dependent functions. Firstly, it can be seen from Figure R1 that the TDC of particles is significantly greater than that of heat, especially in the eastern China. Then, we can also find that the new TDC in the mountain area (i.e., indicated by red dotted rectangle in Figure R1) is smaller than original TDC, because when the whole region is in stable stratification, the complex terrain of the mountain area will disturb the flow field and making the stratification in the mountain area more unstable. With the increase of instability, the new TDC will be smaller than that of the original scheme (Figure 3 in Jia et al., 2021, we mainly compare red ($f_c(Ri)$) and black($f_h(Ri)$) two solid lines). However, compared with the original scheme, the trend of the new TDC is a very good phenomenon for us. We found that the pollutant concentration was not only overestimated in eastern China by the original scheme, but also underestimated in mountainous area of North China. what we expect is that the trend of the new TDC can not only improve the overestimation of pollutant concentration in eastern China, but also improve the underestimation of pollutant concentration in mountainous are of North China. Unfortunately, the results are not as good as we expected. The increasing turbulent diffusion significantly reduces the simulated pollutant concentration in eastern China, but eh decreasing turbulent diffusion does not increase the pollutant concentration in mountainous area of North China. Therefore, on the one hand, this study proves that the main reason for the improvement of pollutant concentration in eastern China is that the new TDC plays a critical role, and puts forward the advantages of the new TDC in the model. On the other hand, we try to explain why the new TDC in mountainous area is insensitive to the simulation of pollutant concentration. We found that other processes may affect the change of pollutant concentration, such as advection transport. The wind speed in mountainous area is relatively high, and the pollutant concentration in the downwind area will be significantly affected when there is an obvious pollutant concentration gradient in the upwind and downwind areas.

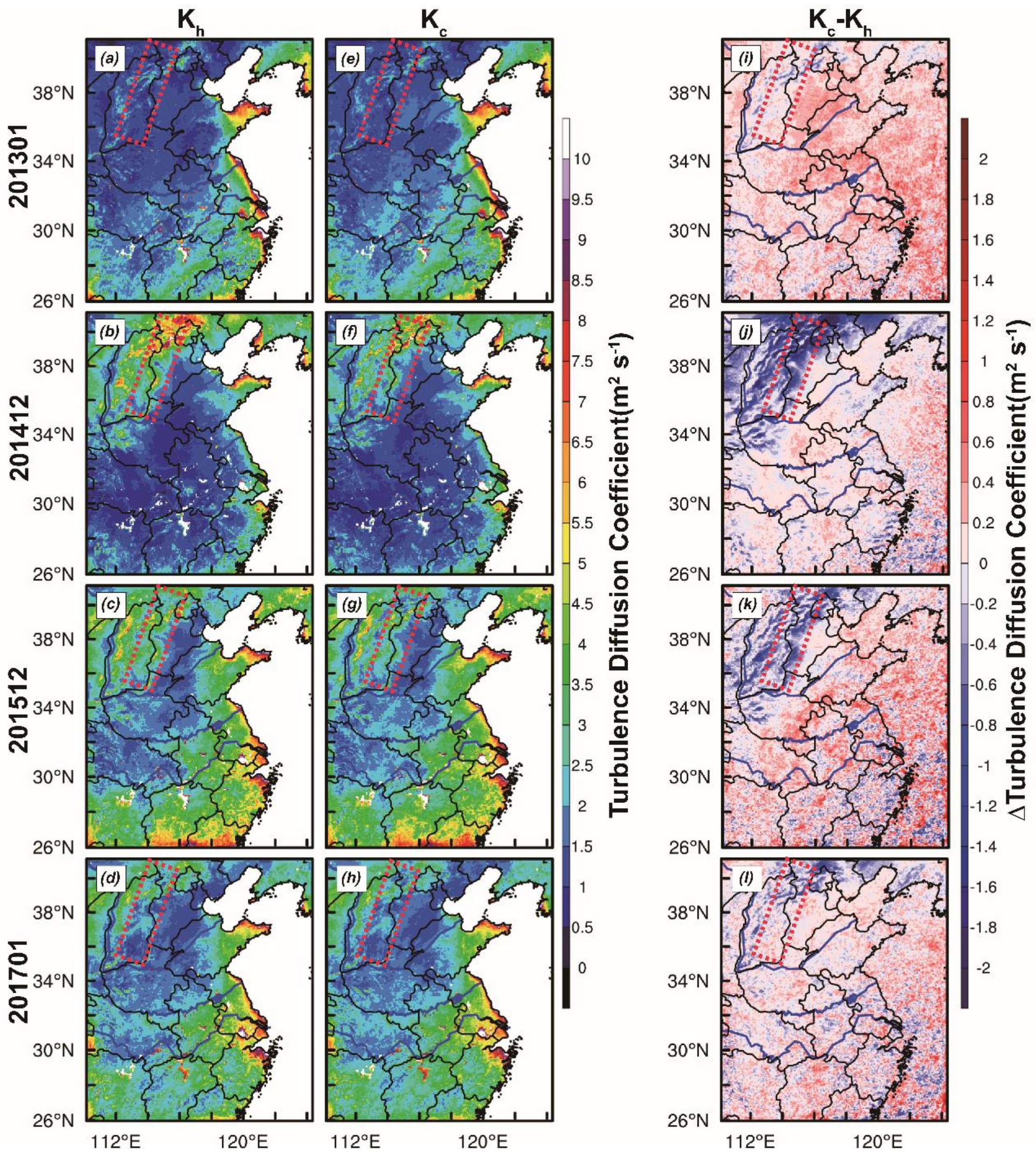

Figure R1. Turbulent diffusion coefficients of (a-d) heat and (e-h) particles, and (i-l) the difference between two turbulent diffusion coefficients.

2) In Section 3, the authors present the analytical results of correlation coefficients R_{wt}, R_{wc} and $R_{wt,wc}$ from observational data. They point out that the behaviors of turbulent transport of heat and particle are different, as shown in Figs. 2b–2d. Thus they use these results to support their choice of a different scheme of turbulent transport for particle from that for heat. In think these results do not make sense. On the one hand, Figs. 2b and 2c show that R_{wt} and R_{wc} are quite different under unstable conditions (i.e., in the daytime), but the authors choose the same scheme of turbulent transport for heat and

particle. On the other hand, the behavior of R_{wc} is not changed under both stable and unstable conditions—the values of R_{wc} fluctuate around zero in the whole day as shown in Fig. 2c, but the authors choose different schemes of turbulent transport for particle under stable and unstable conditions respectively. As for the results show in Fig. 2d, I do not know what the results mean and how to interpret them. In my opinion, Section 3 is not a necessary part for this paper, because the results in this section cannot provide evidence to support the choice of a different scheme of turbulent transport for particle from that for heat under stable conditions. Actually, the authors choose the new scheme according to the results in Jia et al. (2021). They have cited the literature in this paper. That's enough. So I suggest the authors to delete this part, as well as Fig. S1.

Response:

We quite agree with you. According to your opinion, we have deleted this section and enriched the following contents.

3) For the results shown in Fig. 6, the authors state in lines 217-222 "Theoretically, increasing turbulent diffusion will reduce the pollutant concentrations near the surface-layer, and the pollutants will be more fully mixing in the vertical direction, which results in lower concentrations of pollutants in the near surface-layer and higher concentrations of pollutants in the upper layer. Actually, the pollutant concentration is reduced in the surface-layer and it is increased in the upper layer at night (Fig. 6), which is consistent with the theory". If the new scheme has a larger f_c, it is likely that these statements are reasonable. However, the new scheme actually has a smaller f_c. How to interpret the results shown in Fig. 6? Of course, a smaller f_c unnecessarily means a smaller eddy diffusivity K_c. As expressed in Eq. (2), if f_c becomes smaller, while the wind shear becomes much larger, we can still obtain a larger K_c. Does the model using the new scheme predict larger near-surface wind shear?

Response:

Actually, our new scheme has a larger f_c, so the simulation results are in line with the theoretical expectations. Here, the turbulent diffusion coefficient depends on the difference of $f(Ri)$ function, as you mentioned before. The new scheme can not produce larger wind shear, as we said, the new scheme will not change the simulation of meteorological field. Consequently, we mainly explain the first problem clearly, and the following problems can be solved easily.

4) In subsections 5.1 and 5.2 the authors emphasize that the meteorological parameters and PBL height simulated by the new scheme are not changed when compared to those simulated by the original scheme (They provide evidence shown in Figs. S2–S5). They declare in lines 232-233 "noting that the new scheme does not alter the performance of meteorological fields, which is an advantage of the new scheme". They also declare in lines 264-266 "The results of the simulation of pollutant concentration are improved under the similar PBLH, which further demonstrates that the simulation of pollutant concentration is not only controlled by the PBLH". I think these evidence cannot help us to know why the new scheme can reduce the overestimated PM2.5 concentration simulated by the original scheme. Given the unchanged meteorological field as well as PBLH, it seems that the reduced PM2.5 concentration can only be attributed to the new scheme of TDC. However, if the meteorological field and PBLH are really unchanged, the new scheme will have a smaller TDC and should predict larger PM2.5 concentration. So I suggest the authors to provide additional information about the simulation results. These information should tell us what are changed, as well as the relation between these changes and changed PM2.5 concentration.

Response:

Here, we want to make one point: in the current WRF-Chem model, the turbulent diffusion coefficient (TDC) of heat is used by all scalars, that is to say, the turbulent mixing process of heat, water vapor and various pollutants depends on the TDC of heat. Although the previous research is to change the TDC of heat to affect the simulation results of one variable, it should be noted that changing the TDC of heat not only affect the simulation results of one variable, but also of all scalars (Savijärvi et al., 2002; Du et al., 2020; Liu et al., 2021). When the TDC of heat is changed, the simulation results of one variable will be better, but that of other variables will be better. Therefore, we propose to use the TDC of particles to calculate the turbulent mixing process of particle separately. Then, the new scheme improves the simulation results of pollutant concentration without affecting the change of meteorological field. As a result, the final effect is attributed to the difference of turbulent diffusion. As we mentioned earlier, the TDC of the new scheme becomes larger, which makes the simulation results of pollutant concentration lower than that of the original scheme.

5) Following the above comment, the same situation exists in subsection 5.3. I think the discussion about the influence of mountain terrain on the simulated PM2.5 concentration also cannot help us to know why the new scheme can reduce the overestimated PM2.5 concentration simulated by the original scheme. Furthermore, I do not understand the purpose of presenting the CO results in this subsection. Is the TDC for CO the same as that for particle in the new scheme? If not, why can the CO concentration be improved by the new scheme?

Response:

The main reason we mentioned the impact of topography is that some studies found that the pollutant concentration was overestimated in Yangtze River Delta (Du et al., 2020) and that was underestimated in North China Plain (Wang et al., 2018). However, we found these two phenomena at the same time, and also found that the pollutant concentrations in all regions of North China Plain were not underestimated. The pollutant concentration of stations close to Taihang Mountain and Yanshan Mountain was underestimated, while the pollutant concentration of stations far away from mountainous area are still overestimated, especially in Tianjin (Fig. R2). Then, we mainly discuss the influence of topography. Firstly, we found that the TDC of new scheme is smaller than that of original scheme, because of unstable stratification. Theoretically, smaller turbulent diffusion will lead to higher pollutant concentration, so as to improve the underestimation of pollutant concentration around the mountainous area. However, the change of turbulent diffusion in mountainous area has not significantly improved the simulation of pollutant concentration. Therefore, we think that the impact of other processes may be more obvious in mountainous area. Previous studies using the process analysis found that in addition to the largest contribution of emissions and turbulent diffusion, the contribution of advection transport can not be ignored. Thus, we mainly consider the influence of advection transport in Section 5.3 (it has been changed to Section 4.3).

We introduce the simulation results of CO because we consider the secondary transformation of PM2.5 and the influence of the chemical process. When we change the turbulent diffusion, the simulation results of PM2.5 are improved, but whether that of precursor is also improved. However, due to the limitation of observation data, we do not have enough precursor observation data. So we use CO as a representative species of primary pollutants to prove whether the concentration of primary pollutants is also obvious affected by turbulent diffusion.

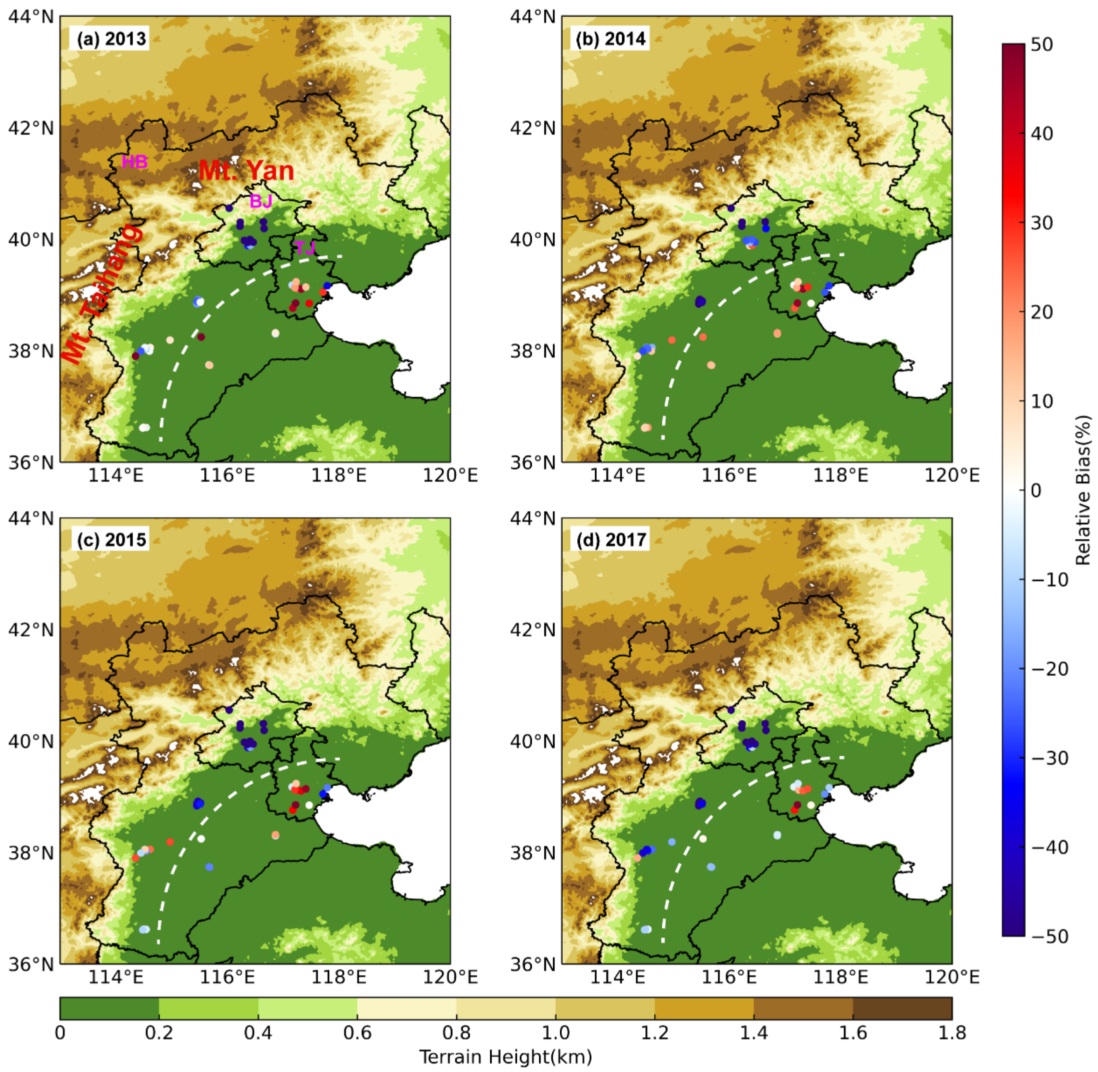

Figure R2. The relative bias (%) between simulation and observation at all environment monitoring stations and terrain height in Beijing-Tianjin-Hebei in (a) 2013, (b) 2014, (c) 2015 and (d) 2017. Taihang Mountain and Yan Mountain are indicated by red text, Beijing (BJ), Tianjin (TJ) and Hebei (HB) are represented by purple abbreviation and the dividing line between overestimated and underestimated areas is indicated by a white dashed line.

I have to say that the paper should be revised substantially. So I think there is no need to give the technical comments. Compared to my major concerns listed above, the technical problems are not important in the present stage.

Response:

Finally, thank you for your valuable comments. According to your comments, we have made substantial changes to the structure and statement of the article.

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