Response to Referee #2 (acpd-14-C5728-2014):

Questions and comments:

The manuscript presents an analysis of an intensive set of measurements of aerosol optical and physical properties from the HaChi experiment in the North China Plain ofthe PRC. The analysis, includes measured data and models based thereon to derive single scattering albedo values as a function of RH and applies the results to an estimate of the NO2 photolysis rate. This is a valuable and important extension to the wealth of data from HaChi that has already been published in the literature and ACP special edition.

Response: Thanks for the comments.

Section 2.2:

The conceptual model of your core-shell mixed aerosol is not clear and needs a better verbal description, mathematical description (see J. Seinfeld and S. Pandis) or illustration.

Given the difference in the models, the last sentence, that the ensemble mean kappa from Liu describes the core-shell mixed aerosol model is not clear. How do these two models, Liu's two-group, externally mixed model, and your internally mixed core-shell model converge? I'm not sure there is an explicit physical, chemical reason for this convergence but I do recall it has been shown to be true empirically for several earlier mixing models. Your core-shell mixing model which seems to allow all degrees of internal, external mixing is more complex.

Response: Thanks for the comments. More information about the core-shell mixing state has been added in section 2.2.

As the referee suggested, we rephrased the paragraph about the two models in section 2.2.3:

"As reported by Liu et al. (2011), two groups of particles can be distinguished based on their hygroscopicity, i.e. the nearly-hydrophobic particles and the more-hygroscopic particles. In detail, the nearly-hydrophobic particles are composed mainly by the LAC and the primary organic aerosol (POA). In this study, the ambient aerosols are classified based on their mixing state, which is quite different from Liu et al. (2011). The core-shell mixed aerosol is composed of not only more-hygroscopic compositions, such as inorganic salts and acids, but also nearly-hydrophobic compositions, such as POA and LAC. The hygroscopic behavior for the core-shell

mixed aerosol can not be represented definitely by Liu et al. (2011). However, as shown in the work of Liu et al. (2014), the less-absorbing component consists the majority (from 80% to 90%) of aerosol populations at all sizes. As a result, the size-resolved number fractions of the core-shell mixed aerosol are higher than 94%, and much larger than the externally mixed LAC. So is the hygroscopicity of the core-shell mixed aerosol. Therefore, the hygroscopicity of the core-shell mixed aerosol is much close to that of the aerosol population. The ensemble mean κ for all groups in Liu et al. (2011) can describe the hygroscopicity of the core-shell mixing particles and used in the calculation of g(D_p,RH)."

Page 7, line 23

Do you mean g rather than f?

f appears in equation 7 and needs to be defined.

r also.

Response: Thanks for the correction. We changed *f* into *g*. Definitions of *r* and *f* can be found in equation (1) and equation (4), respectively.

3.1 Overview

In your discussion it would be useful to refer to the excellent summary data in your table as well as to the figures. The tables are quantitative. Consider adding two more rows for each parameter giving the 10 and 90 percentile. Then, in the text you can refer quantitatively to the percent of the data within those percentages (or within one or two sigma of the mean) rather than "mostly" e.g. page 10 lines 21 and 22 and elsewhere..

Response: Thanks for the suggestion. Boxes and whiskers in figure 2 represent the 5, 25, 75 and 95 percentiles. We have replaced *"mostly"* with these percentages in the manuscript.

You mention correlation of RH and scattering and absorption coefficients. It is difficult to see these by eye, ocular analysis, in your plots. A calculation of correlation coefficient and table would be useful if they are significant, and, or significantly different.

Response: We agree with the referee's comment. In this paper, we focus on the

variation of ambient ω with RH and ω at dry state. The discussion of the correlation between RH and scattering and absorption coefficients has been removed from this manuscript. The correlation between scattering and RH has been discussed in the paper of Chen et al (2012).

3.2

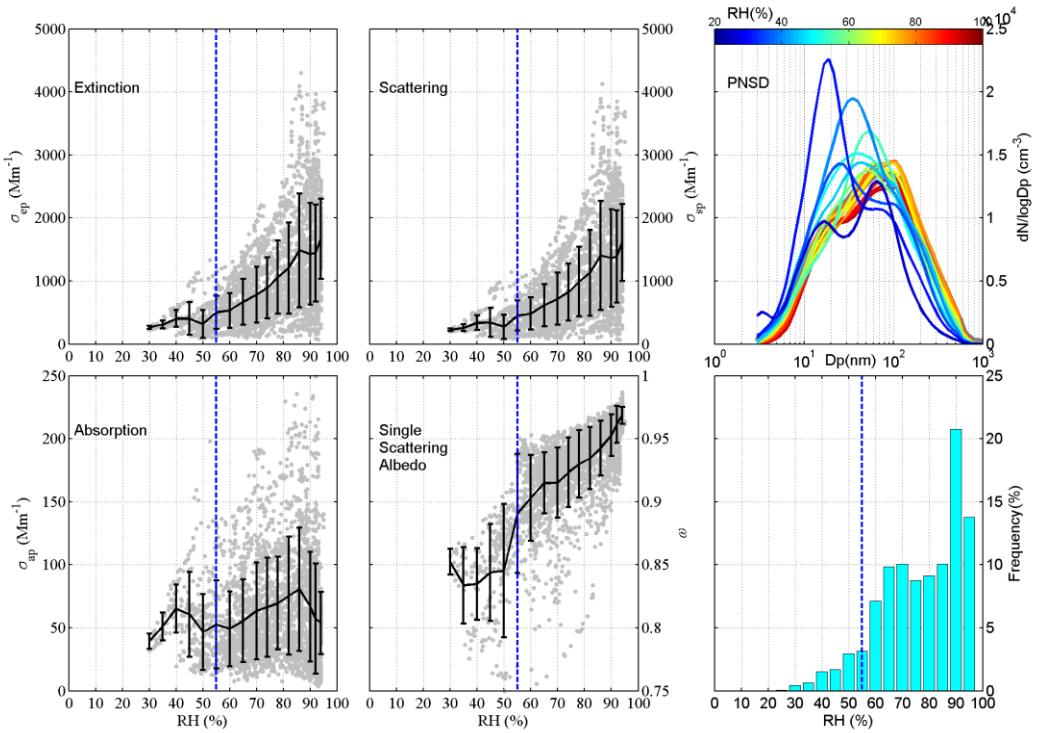
The decrease in scattering at about 85% RH that you show is not observed in the earlier literature nor expected based on models and theory. What is the reason for this?

Regarding Figure 3, the most common way of presenting hygroscopic growth of geometric diameter or optical properties based on RH dependent measurements, is to normalize them with reference to the low RH measurements. This may not be possible in your case if there was no relatively continuous measurement of low RH properties to match the elevated RH measurements and if atmospheric conditions changed during the RH scan period of the HTDMAs. You do have a continuous record of scattering from the nephelometer at 30% RH to test atmospheric time variability of scattering.

Response: Thanks for the comment.

Aerosol scattering coefficient highly depends by the PNSD. In the following figure, the averages of PNSD at different RHs aerosol pollution are shown. The averaged PNSD at 80% RH (orange line) is slightly higher than that at 90% RH (red line), which results in the decrease in scattering at about 85% RH.

The data at RHs less than 55% along with the average PNSD and frequency at different RH are presented in the following figure. At RHs less than 55%, the frequency of measurements is low. So the average of AOP may be affected by individual pollution episode and is not representative in the NCP. The average PNSDs at RHs less than 55% (cooler lines) fluctuate widely, revealing poor representativity. As a result, the ω values drop down and deviate significantly. These data were deleted in our analysis.



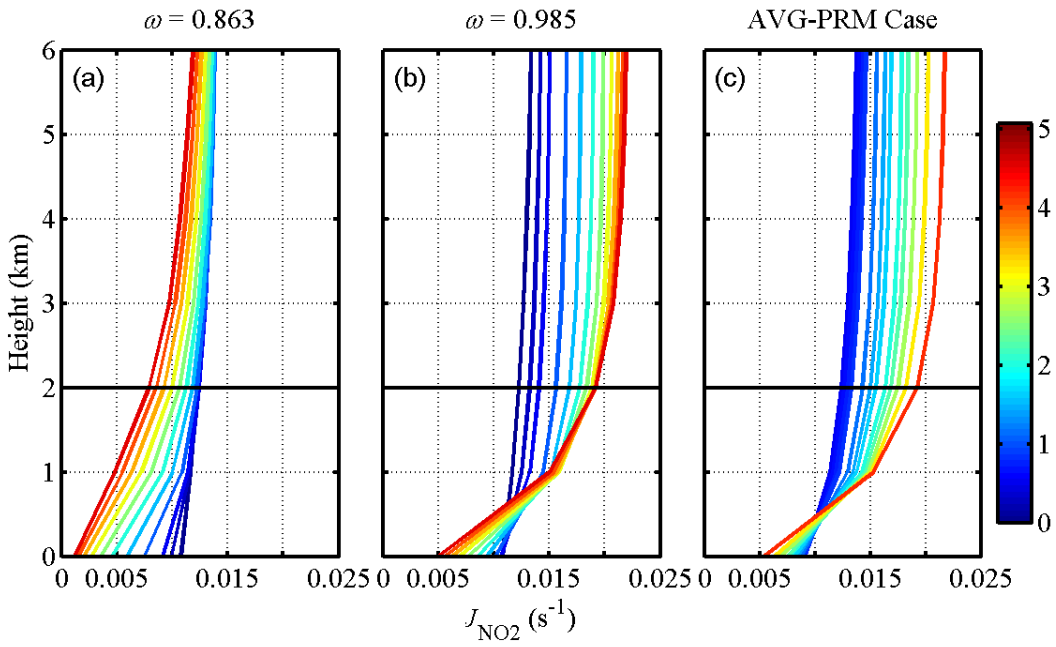
AVG-PRM needs to be explained when first used rather than later.

Response: Thanks for the suggestion. We have added the description as follows:

"This case will be proved to be representative in the NCP by the analysis in Fig. 4, and is compared with the ambient AOP here."

The numerical labels on the x-axes are confusing. Put them all below the individual plots. The plots would be easier to read and compare if all the JNO2 scales were the same, i.e., 0 to 0.25.

Response: Thanks for the suggestion. We have revised the figure and its description.



Technical errors and suggestions:

Abstract

Line 16

aerosol hygroscopic growth on ~~the~~ ω and its application ~~on~~ to the NO2 photolysis rate coefficient (JNO2) are

Response: Thanks for the suggestion. We have revised it accordingly.

Line 22

by a dry state

Particles are seldom really dry, anhydrous. I prefer to use the qualifier, "low, < xx% RH" where xx is often a value of about 30% but could be <10%. Once defined for the instrumental system, further use of "dry state" is OK.

Response: Thanks for the suggestion.

Line 23

~~The~~ A Monte Carlo simulation shows that the uncertainty of ω from the propagation of uncertainties in the input parameters decreases from 0.03 (at dry state) to 0.015 (RHs > 90%).

This is to be consistent with 0.015 as stated on page 15.

Response: Thanks for the suggestion. We have revised it accordingly.

Page 2
Line 3

photolysis of NO2 at the ground level, whereas accelerates it above the ~~upper~~ moist planetary boundary layer.

Response: Thanks for the suggestion. We have revised it accordingly.

Line 12

aerosol's radiative effect ~~as a~~and is a significantly uncertain factor. Defined as the ~~relative magnitude~~ratio of absorption to the sum of ~~and~~ scattering plus absorption,

Response: Thanks for the suggestion. We have revised it accordingly.

Line 15

~~At~~ Under dry conditions (< xx% RH),
the ~~relative~~ complex refractive index

Response: Thanks for the suggestion. We have revised it accordingly.

Line 18

is highly ~~varied~~variable

Response: Thanks for the suggestion. We have revised it accordingly.

Line 26

Aerosol absorption ~~slightly~~ varies slightly with RH, and is often considered to be constant. (reference, see Beni Brem, PhD thesis Univ. of Illinois. Measured Optical Properties of Inorganic and Organic Aerosols at Relative Humidities up to 95% Benjamin T. Brem, Francisco C. Mena Gonzalez, Scott R. Meyers, Tami C. Bond, Mark J. Rood Aerosol Science and Technology, 46: 178-190, 2011) Thus, the value of w can be RH~~s~~ dependent and

Response: Thanks for the suggestion. We have revised it as follows:

Aerosol absorption is often considered to vary slightly with RH, while Brem at al. (2012) report the enhancement of aerosol absorption at high RH. Thus, the value of ω can be RH dependent and…

Page 3
Line 1

~~As~~ Because ~~the~~ ambient air is most often sampled in ~~sheltered conditions~~a shelter or structure ~~in most of the aerosol measurements~~, it's very important to measure and report the RH at the point of measurement and to apply ~~the~~ coincident measurement of aerosol hygroscopicity (or a model thereof) to ~~quantifying~~ quantify the ambient w (Nessler et al., 2005).

Response: Thanks for the suggestion. We have revised it accordingly.

Line 4

Due to the high sensitivity of radiative forcing to the variation of w, it is essential to obtain ~~realistic~~ atmospherically relevant values of w

Response: Thanks for the suggestion. We have revised it accordingly.

Line 9

RHs can strengthen the forcing by ~~several times~~a factor of two or more

Response: Thanks for the suggestion. We have revised it accordingly.

Line 11

depth (t) are the relevant parameters in the

Response: Thanks for the suggestion. We have revised it accordingly.

Line 14

~~The s~~Sensitivity ~~study~~ studies shows~~s~~ that RH~~s~~

Response: Thanks for the suggestion. We have revised it accordingly.

Line 17

The North China Plain (NCP) with several megacities and the location of plenty of ~~industries~~ industry ~~located~~ suffers ~~a series 17 of~~frequent severe aerosol pollution episodes

Response: Thanks for the suggestion. We have revised it accordingly.

Line 24

growth factors at RHs up to 98.5% measured by a High Humidity Tandem Differential Mobility Analyzer (HH-TDMA) ~~instrument~~ indicated the existence of a dominant more-hygroscopic group of

Response: Thanks for the suggestion. We have revised it accordingly.

Line 27

and ~~assisted~~ in ~~the~~ combination ~~between~~ contributed to the enhancement of extinction at high RHs and the low visibilities ~~for~~ on hazy days

Response: Thanks for the suggestion. We have revised it accordingly.

Page 4

Lines 1 and following

at high aerosol condition is still ~~uncovered~~unresolved

Response: Thanks for the suggestion. We have revised it accordingly.

In this study, the RH ~~dependent~~ dependence of aerosol optical properties are ~~re~~presented and their influences on UV radiation are investigated.

Response: Thanks for the suggestion. We have revised it accordingly.

The descriptions of data, calculations and models are presented in ~~Sect.~~section 2~~.~~ ~~;~~ ~~The~~ overviews of aerosol optical properties are in section~~Sect~~. 3~~.~~ ~~;~~ ~~The~~ results of modeled UVB irradiance and NO2 photolysis rate coefficient (JNO2) are represented in section~~Sect~~. 4~~.~~ ~~;~~ ~~There~~ and there is ~~the~~ a summary in section~~Sect.~~ 5.

Response: Thanks for the suggestion. We have revised it accordingly.

Line 20

~~Detailed~~ Details ~~information~~ of aerosol, radiation and meteorological parameters ~~was~~ were investigated
The measurements were ~~preformed~~ performed in an aerosol sampling container that was ~~at~~ maintained at a temperature of

Response: Thanks for the suggestion. We have revised it accordingly.

Line 24

samples were dried by an automatic aerosol diffusion dryer to keep the sample RH ~~below~~ less than 30 %. Further information ~~of~~ regarding aerosol measurements is documented in the relevant studies on aerosol in the HaChi project

Response: Thanks for the suggestion. We have revised it accordingly.

Line 27

Particle number size distribution (PNSD) in the range of 3 nm - 10 um was determined ~~jointly~~ by the combination of an Aerodynamic Particle Sizer (APS Model 3321, TSI, Inc., Shoreview, MN USA) and a Twin Differential Mobility Particle Sizer

Response: Thanks for the suggestion. We have revised it accordingly.

Page 5
Line 7

~~Scattering~~ Particulate scattering coefficients at the wavelengths of 450, 550 and 700 nm were ~~observed~~ measured by ~~the~~ an integrating nephelometer (Model 3563, TSI, Inc., Shoreview, MN USA~~, Model 3563~~

Response: Thanks for the suggestion. We have revised it accordingly.

The geometric hygroscopic growth factor (g(RH,Dp)) ~~at RHs from 0% and 98%~~relative to the dry state was obtained from ~~the~~ measurements of the High Humidity Tandem Differential Mobility Analyzer (HH-TDMA). The HH-TDMA

measured ~~Growth Factors~~ (g(RH,Dp)) for particles ~~of~~ in four selected diameter increments (at 50 nm, 100 nm, 200 nm and 250 nm) at three RHs (90 %, 95 % and 98.5 %) with an absolute accuracy of ±1.2% for 98%.

Response: Thanks for the suggestion. We have revised it accordingly.

Do you mean 98% or 98.5% for the accuracy statement?

Response: Thanks for the comment. It should be 98.5% and we have revised it accordingly.

Line 13

~~In~~ At the Tieta site, the CE-318 Sun-photometer was used to measure the aerosol optical depth at ~~the~~ four wavelengths of 440nm, 670nm, 870nm and 1020nm

Response: Thanks for the suggestion. We have revised it accordingly.

Line 18

~~1~~ One minute~~min~~ data of meteorological parameters

Response: Thanks for the suggestion. We have revised it accordingly.

Line 23

~~Dependence~~ the RH dependence of w and other aerosol optical parameters can be calculated using the averaged PNSD 23 and the Mie code (BHCOAT),

Response: Thanks for the suggestion. We have revised it accordingly.

Give a reference here for this MIE BHCOAT code that is presented later.

Response: Thanks for the suggestion. We have added the reference.

Page 6

Line 4

aerosols chemical components are divided into two classes based on their refractive indices, i.e.1) the LAC and 2) the less absorbing components (inorganic salts and acids, and most of the organic compounds). ~~Two~~ Within these classes two types of particles are assumed:

Response: Thanks for the suggestion. We have revised it accordingly.

Line 14

Under the assumption in sect. 2.2.1, total PNSD of the aerosol population is comprised of subsets PNSDs of the

Response: Thanks for the suggestion. We have revised it accordingly.

Page 9
Line 11

the sum of those two classes. The aerosol extinction coefficient ~~comes~~ (σ_{ep}) ~~defines~~ is defined as $\sigma_{ep} = \sigma_{sp}+\sigma_{ap}$, and ω is ~~defines~~ defined as $\omega = \sigma_{sp}/\sigma_{ep}$~~, as expected~~.

Response: Thanks for the suggestion. We have revised it accordingly.

Line 15

model with eight-stream discrete ordinate solver. ~~Giving~~ Given the information of aerosol and cloud optical properties, this model can calculate spectral irradiance ~~at~~ over wide range of wavelengths (121nm~735nm) and photolysis rate coefficients of important photochemical reactions in atmosphere at specific location and time. In this paper, irradiance at the wavelengths from 280nm to 320nm is calculated and compared with observations. JNO2 is used to investigate the influence of aerosol ~~of~~ on ozone photochemistry.

Response: Thanks for the suggestion. We have revised it accordingly.

Line 24

introduced in Sect. 2 for ~~with~~ the dataset measured during HaChi summer campaign. The aerosol

Response: Thanks for the suggestion. We have revised it accordingly.

Page 10
Line 2

in Fig. 1. For the majority of the observation, the 1 minute wind speed is mostly ~~lower~~ less than

Response: Thanks for the suggestion. We have revised it accordingly.

Line 3

The overall σ_{sp} is ~~among~~ within the

Response: Thanks for the suggestion. We have revised it accordingly.

Line 15

~~On the~~By contrast, for example on July 16th, July 22nd and August 13th, with typical wind speed (about 3m/s) and ~~extremely~~ relatively low RH (about 50%), the decrease of ~~the~~ σ_{sp} was stronger than ~~the~~ that of σ_{ap}, and ω reached the low values of 0.75, 0.7 and 0.75, respectively, on those three days. The value of ω ~~is~~ was not sensitive to the direction of the wind ~~speed as well~~.

Response: Thanks for the suggestion. We have revised it accordingly.

Line 21

maximum at about 06:00 ~~am~~ and a minimum at about 16:00 ~~am~~. Values of σ_{sp} ranged mostly from about 500

AM and PM are not needed if you use a 24 hour time scale as you do in the figure.

Response: Thanks for the suggestion. We have revised it accordingly and changed the *"am"* to *"LT"*.

Line 25

~~outstanding~~clear diurnal patterns are ~~verified by~~consistent with the high values

Response: Thanks for the suggestion. We have revised it accordingly.

This ~~conspicuous~~ obvious diurnal

Response: Thanks for the suggestion. We have revised it accordingly.

The diurnal patterns of the ambient σep, σsp and σap are similar ~~with~~ to their diurnal patterns ~~at~~ in the dry state

Response: Thanks for the suggestion. We have revised it accordingly.

However, as Ma et al. (2011) reported, ω ~~at~~ in the dry state reaches its peak at noon and ~~gets its bottom in~~a minimum in the ~~nearly~~ morning and again in the evening

Response: Thanks for the suggestion. We have revised it accordingly.

page 11
line 22

The absorption coefficient of the aerosol in the NCP seems to be independent ~~from~~ of RH in Fig. 3c, which is expected according with Pan et al (2009). Therefore, the value of ω ~~at ambience~~ under ambient conditions can be estimated from the two

Response: Thanks for the suggestion. We have revised it accordingly.

As expected, the ω approaches higher values

Response: Thanks for the suggestion. We have revised it accordingly.

Page 12
Line 1

The distribution of ω_0 is mainly ~~distributes~~ in the range of 0.80 to ~~~~0.95 and has an average of about 0.863.

I don't think three significant figures are justified.

Response: Thanks for the comment. We have revised it accordingly.

Considering that over half of the ω0 ~~is among~~values are in the range 0.85~0.9, the value of 0.863 is representative ~~in~~ for the NCP

Response: Thanks for the suggestion. We have revised it accordingly.

Page 13
Line 2
decreases with ~~the~~ increasing ~~of (~~secant of~~)~~ solar zenith angle

Response: Thanks for the suggestion. We have revised it accordingly.

page 15
line 2
~~As an implication~~A derived parameter, the

Response: Thanks for the suggestion. We have revised it accordingly.

Line 5
The hygroscopic growth influences not only aerosol PNSDs but also the refractive index of the aerosol. In this study, the shell of the core-shell mixed aerosol is assumed to be composed of ~~the~~ less absorbing, water soluble components with the refractive index of 1.53–10−7i. As the ambient RH increases, ~~liquid~~ water vapor is ~~un~~taken up by the shell and the less absorbing components ~~get dissolved~~dissolve to maintain water vapor equilibrium between the ambient air, and the liquid shell. The refractive index of the shell is determined by the water content and the ~~solution~~ solute together

Response: Thanks for the suggestion. We have revised it accordingly.

Table 1
Give units within parentheses e.g., (Mm^{-1}) rather than after a slash which looks the same as the mathematical symbol for 'divided by'.

Response: Thanks for the suggestion. We have revised it accordingly.

Table 2
Since you give the input uncertainties here, it would be good to list the major output uncertainties also.

Response: Thanks for the suggestion. The output uncertainties are listed in Table 3. It will be too complicated to combine these two tables.

Table 3 caption

Table 3. The standard deviation of ω (σ_ω), the ~~growth rate~~rate of change of ω with

RH~~s~~

Response: Thanks for the suggestion. We have revised it accordingly.

Fig. 7. Altitude profiles of JNO2 in three cases (a, b, c) the same ~~with~~ as Fig. 6. Colors represent the value of t, as expected. The lower plots show the profiles in 2km (d, e, f).

Response: Thanks for the suggestion. We have renewed the Figure 7 and revised the description as follows:

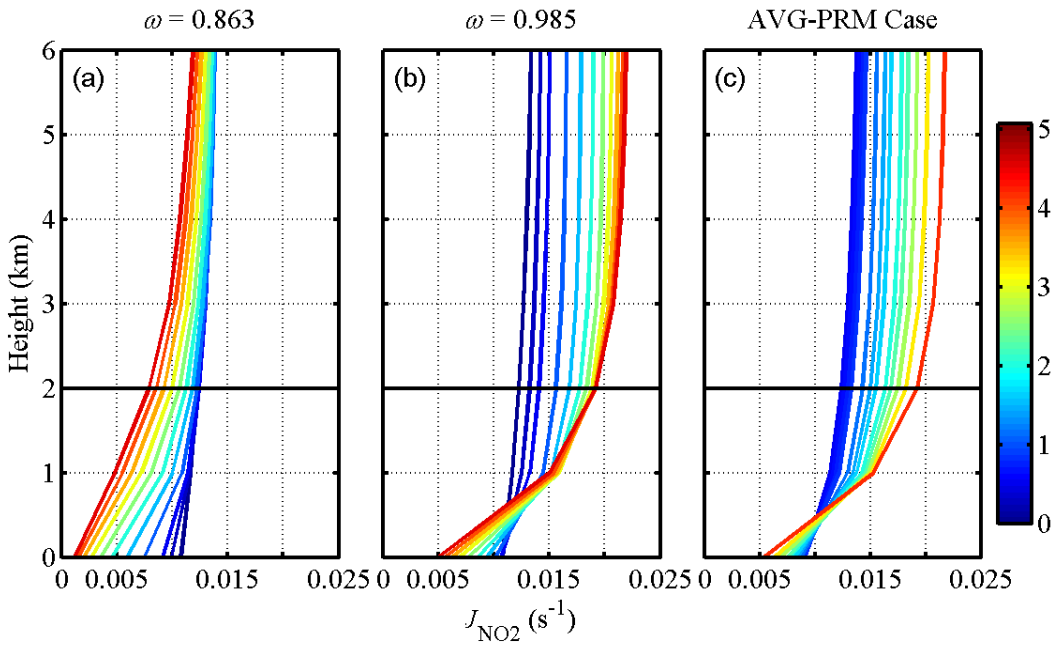


Fig. 1. Altitude profiles of J_{NO2} in three cases (a, b, c) the same with Fig. 6. Colors represent the value of τ, as expected. The black lines lie on the height of 2 Km.

Fig. 8. The ~~growth rate~~increase of JNO2 with ~~as~~ t ~~goes on in~~for three cases: ω ~~is~~ = 0.985 (circle), ω ~~is~~ = 0.863 (square) and ~~2~~an RH-dependent ω (AVG-PRM case, triangle).

Response: Thanks for the suggestion. We have revised it accordingly.

Reference:

Brem, B. T., Mena Gonzalez, F. C., Meyers, S. R., Bond, T. C., and Rood, M. J.: Laboratory-measured optical properties of inorganic and organic aerosols at relative humidities up to 95%, Aerosol science and technology, 46, 178-190, 2012.

Chen, J., Zhao, C. S., Ma, N., Liu, P. F., Gobel, T., Hallbauer, E., Deng, Z. Z., Ran, L., Xu, W. Y., Liang, Z., Liu, H. J., Yan, P., Zhou, X. J., and Wiedensohler, A.: A parameterization of low visibilities for hazy days in the North China Plain, Atmospheric Chemistry and Physics, 12, 4935-4950, 10.5194/acp-12-4935-2012, 2012.

Liu, H. J., Zhao, C. S., Nekat, B., Ma, N., Wiedensohler, A., van Pinxteren, D., Spindler, G., Müller, K., and Herrmann, H.: Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China Plain, Atmos. Chem. Phys., 14, 2525-2539, 10.5194/acp-14-2525-2014, 2014.

Liu, P., Zhao, C., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W., Deng, Z., Ma, N., and Mildenberger, K.: Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain, Atmos. Chem. Phys, 11, 3479-3494, 2011.