Comment from Referee #2:

The authors use the WRF-Chem model to examine the effect of a total solar eclipse on atmospheric pollutants occur over China. The overall idea is excellent and the scientific results and conclusions are presented in a clear, concise, and well structured way. However, the scientific quality, relative to the reproduction of the radiation change due to the solar eclipse, is quite poor and this affects strongly the results and the scientific value of the manuscript. The authors try to reproduce the solar eclipse by varying the solar constant and the photolysis rates to the same extent. The variations are introduced by the use of a scaling factor dependent on latitude, longitude and time. Then, they try to validate this hypothesis with downward solar radiation measurements, for which, it is true, the variation during the time of the eclipse is approximately proportional to the obscuration of the sun. However, it has been shown clearly in recent studies (both in ACP, Emde and Mayer(2007), Kazantzidis et al. (2007) and references therein), that the effect of the eclipse on surface irradiance has spectral characteristics due to the limb darkening. According to those studies, the reduction (relative to non-eclipse conditions) in UVB is almost 1.5 higher than UVA and visible. This effect should be introduced in the WRF-Chem radiation scheme, since it affects significantly the photochemical calculations of this study. In this case, most of the scientific work and analysis should be redone. So, I recommend the rejection of the manuscript.

Reply to Referee #2:

The authors thank the Referee for reviewing our paper and giving valuable comments. In our original study, the reproduction of the solar eclipse is introduced by the use of a scaling factor dependent on latitude, longitude and time. However, some previous studies (Emde and Mayer, 2007; Kazantzidis et al., 2007 and references therein) showed that the effect of the eclipse on surface irradiance has spectral characteristics due to the limb darkening, which leads to more pronounced decrease in the radiation

at the lower wavelengths. Therefore, it is more precise to take the limb darkening into account, and the mathematics of the limb darkening and its effect on different wavelengths during the solar eclipse are introduced in Section 1 of this reply. Furthermore, the authors decided to study the sensitivity analysis of limb darkening effect on surface ozone using a box model (Section 2), which is also added into the revised manuscript. Finally, we conduct the WRF-Chem simulations with a corrected scaling factor which is dependent on latitude, longitude, time and wavelengths:

J = J * fac(lat,lon,time,wavelength)

The reproducing the solar eclipse in the WRF-Chem model was solved by varying the solar radiation and photolysis rates according to the obscuration of the sun, which alters by different grid point and time. For example, the obscuration of the sun in each grid point is proportional to the distance from the center of the total eclipse at a given time. Since the moon's umbral shadow moved with a specified velocity on earth, the obscuration in each grid point needs to be rescaled in the next time step. In addition, the limb darkening effect is introduced in this scaling factor, which shows that shorter wavelength radiation reduced slightly more at a given obscuration. The renewed discussion can be referred to the revised manuscript.

1. Mathematics of limb darkening

During different times of an eclipse, the disk of the moon covers different parts of the limb and the center of the sun. The radiation from the limb of the solar disk is less intense than that from the centre. Moreover this limb darkening is spectrally dependent. Thus the spectral composition of the irradiance of the sun changes during the eclipse (Emde and Mayer, 2007; Kazantzidis et al., 2007; Koepke et al., 2001).

Based on considerations of the radiative transfer within the solar atmosphere, the limb darkening can be expressed as a function of r, the relative distance from the centre of the solar disk (Waldmeier, 1941; Scheffler, 1974),

$$\Gamma_\lambda(r) = \frac{I_\lambda(r)}{I_\lambda^0} + \frac{1+\beta_\lambda \cdot \sqrt{1-r^2}}{1+\beta_\lambda}$$

(E   1   )

Where r=0 represents the centre and r=1 the limb of the sun. $I_\lambda^0=I_\lambda(r=0)$ denotes the intensity coming from the centre of the solar disk.

The limb darkening coefficient β_λ is a function of wavelength (Waldmeier, 1941):

$$\beta_\lambda = \frac{3 \cdot h \cdot c \cdot \sqrt[4]{2}}{8 \cdot k \cdot \lambda \cdot T_S}$$

(E   2   )

Where λ is wavelength in m, h the PLANCK constant ($6.63 \cdot 10^{-34} Js$), c the speed of light ($2.998 \cdot 10^8 ms^{-1}$), k the Boltzmann constant ($1.38 \cdot 10^{-23} JK^{-1}$) and Ts the temperature of the sun's surface (5740K).

Using $\Gamma_\lambda(r)$ given in E1, the spectral irradiance of the uncovered sun can be calculated by integrating over the whole solar disk (Koepke et al., 2001):

$$I_{s\lambda} = \int_0^{Rs} \int_0^{2\pi} I_\lambda^0 \cdot \Gamma_\lambda(r) \cdot r \cdot d\alpha \cdot dr = 2\pi \cdot I_\lambda^0 \int_0^{Rs} \Gamma_\lambda(r) \cdot r \cdot dr \quad (E3)$$

During the eclipse, part of the solar disk is covered by the moon leading to a reduced solar intensity $I_{c\lambda}$. The distance between the centres of the disks of the moon and the sun is denoted as X, which is linearly correlated to time. When X>Rm and r≤X-Rm the angular integration is performed over a complete circle. When X<Rm and $r \leq Rm - X$ the annulus is behind the disk of the moon and omitted from integration. For $|X - Rm| < r$ a sector with an angle 2α will not contribute to the radiation and

will be excluded from integration. This angle α can be determined from geometrical considerations as a function of X and r (Koepke et al., 2001):

$$\alpha(r,X) = \begin{cases} 0, & \text{if } X - Rm \geq r \text{ and } X > Rm \\ \arccos\left[\frac{r^2 + X^2 - Rm^2}{2 \cdot r \cdot X}\right], & \text{if } |X - Rm| < r \\ \pi, & \text{if } Rm - X > r \text{ and } X \leq Rm \end{cases} \quad (E4)$$

The integration of the irradiance from the remaining solar disk not covered by the moon thus becomes (Koepke et al., 2001):

$$I_{c\lambda} = \int_0^{Rs} \int_0^{2\pi - \alpha} I_\lambda^0 \cdot \Gamma_\lambda(r) \cdot r \cdot d\alpha \cdot dr$$

$$= 2\pi \cdot I_\lambda^0 \int_0^{Rs} \left[1 - \frac{\alpha(r,X)}{\pi}\right] \Gamma_\lambda(r) \cdot r \cdot dr \quad (E5)$$

This equation has been integrated numerically. For a description of the spectral variation of the solar irradiance during eclipse, the normalized irradiance I_{norm}, i.e. the ratio between the irradiance of the remaining part of the sun $I_{c\lambda}$ (E5) and the irradiance of the uncovered solar disk $I_{s\lambda}$ (E3), is used:

$$I_{norm} = \frac{I_{c\lambda}}{I_{s\lambda}}$$

(E 6)

Fig. 1a shows the decrease in I_{norm} with decreasing X due to obscuration of the sun covered by the moon. The bold grey line represents the normalized intensity of a hypothetical sun without limb darkening (noLD). $I_{norm}(\lambda)$ is wavelength dependent

because of limb darkening. These wavelengths are chosen to cover the spectral range of interest. 310nm is important for the photochemical processes of O_3 and NO_2. The wavelength 550nm is given as a standard wavelength for visible light and 1500nm is shown as an example for a wavelength of the infrared spectral range. Compared to the line of no limb darkening (noLD), the normalized radiation for each wavelength ($I_{norm}(\lambda)$) enhances when the distance of the centers of moon and sun is larger than 0.5 ($|X|>0.5$). During that time when limb of the sun is covered by the moon, the brighter central region of the sun still emits large radiation. During the central parts of the sun obscured by the moon ($|X|<0.5$), $I_{norm}(\lambda)$ reduces because larger radiation in these region is blocked.

Fig. 1b presents the spectral dependence of the solar radiation during the eclipse when compared to the case of no limb darkening (noLD), which is calculated by formula:

$$\{I_{norm}(\lambda) - I_{norm}(noLD)\}/I_{norm}(\lambda) * 100\%$$

$$(E7)$$

It is obvious that shorter wavelength is affected more by limb darkening effect. The reduction of the radiation is more significant when close to the central part of the eclipse. When $|X|>0.9$, the $I_{norm}(\lambda)$ is reduced by more than 60%, 30% and 10% at 310 nm, 550nm and 1500nm respectively.
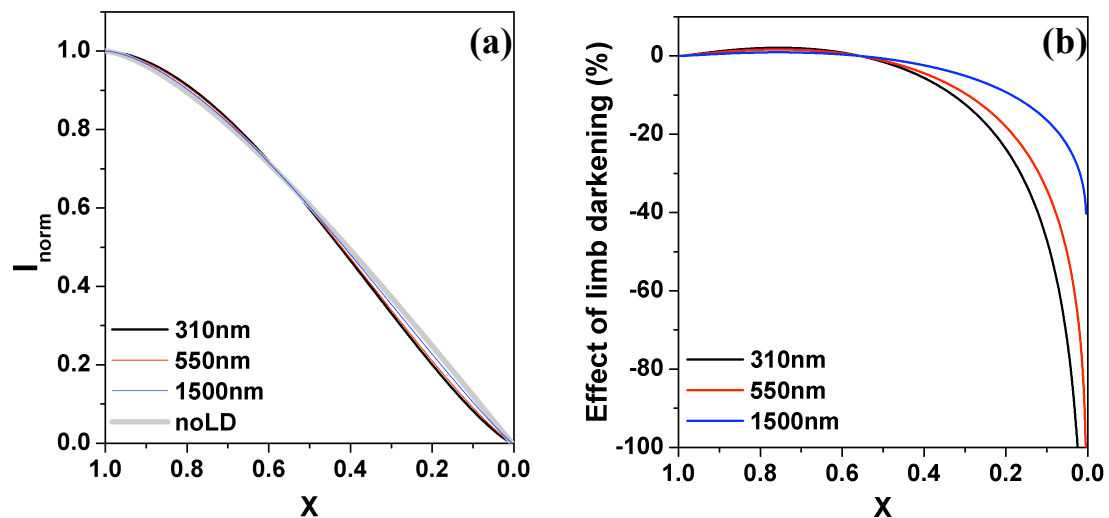
Fig. 1 Normalized radiation as function of X, the distance of the centres of moon and sun, during eclipse for different wavelengths (a); Effect of limb darkening as a function of X for different wavelengths, the calculation formula is:

$$\{I_{norm}(\lambda) - I_{norm}(noLD)\}/I_{norm}(\lambda) * 100\%$$

2. Model uncertainties from limb darkening effect

As mentioned in Section 1, the spectral dependence of radiation due to the limb darkening of the sun becomes relevant during a solar eclipse. It is necessary to investigate how the limb darkening affects the surface ozone. A CBM-Z chemical box model (Zeveri et al., 1999) is used to carry out three sensitivity experiments, which is depicted in table 1.

Table 1 description of sensitivity experiments

case	description
Non-Eclipse	Normal radiation according to solar zenith angle
Eclipse-LD	Reduced radiation with limb darkening effect (black line in Fig. 1a)
Eclipse-noLD	Reduced radiation without limb darkening effect (grey line in Fig. 1a)

It is known that 310nm is important for the photochemical processes of O_3 and NO_2. And the photolysis rate is directly influenced by solar radiation at this wavelength. Therefore, the variation of photolysis rate can be factored by the reduction of $I_{norm}(310nm)$ showed in Fig. 1a. It should be noting that Fig.1a presents half part of an eclipse, and there is another part of eclipse which denotes that the moon leaves the sun gradually. Fig. 2 shows the Photolysis rate of ozone (JO^1D, s^{-1}) in three sensitivity experiments during a solar eclipse event. The JO^1D value in Non-Eclipse experiment

displays steadily increase in the morning time, while Eclipse-LD and Eclipse-noLD experiments reveal reduction during the eclipse period. The difference of JO^1D between Eclipse-LD and Eclipse-noLD is the same extent as $I_{norm}(\lambda)$ between 310nm and noLD (Fig. 1a).

In Kazantzidis (2007)'s work, also cited by the Referee, Radiative transfer model results were used for the computation of a correction for the total ozone measurements due to the limb darkening. This correction was found too small (less than 0.01%) to explain the large decrease in total ozone column derived from the standard Brewer measurements. Here we confirm this find by the ozone concentration from the box model (Fig. 3). Ozone concentration in Non-Eclipse run shows steady increase during the solar eclipse period, while Eclipse-LD and Eclipse-noLD results present a slightly decrease. It is worth-noting that these two lines (Eclipse-LD and Eclipse-noLD) are almost the same, with more pronounced decrease in Eclipse-LD experiment. The effect of limb darkening on surface ozone is less than 0.5%. Although the effect of limb darkening on shorter wavelength is larger (Fig. 1b), the absolute difference of reduced solar radiation between short and long wavelength is small (Fig. 1a). This small perturbation leads to limited change in surface ozone between Eclipse-LD and Eclipse-noLD experiment. Therefore, it is concluded that the effect of limb darkening on surface ozone is too small to explain the large decrease in surface ozone during a solar eclipse event.
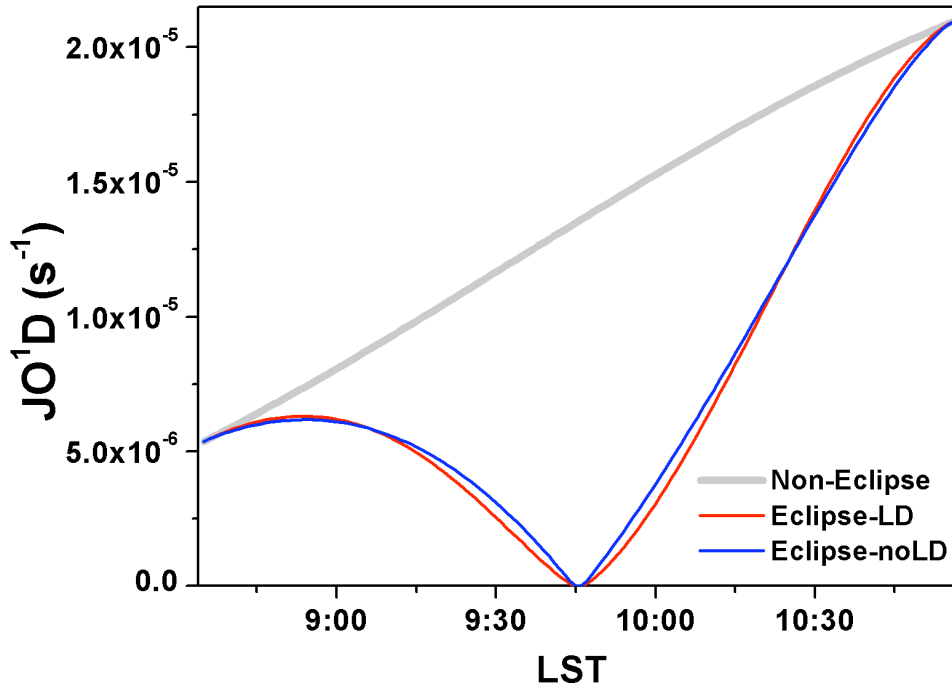
Fig. 2 Photolysis rate of ozone (JO^1D, s^{-1}) in three sensitivity experiments.
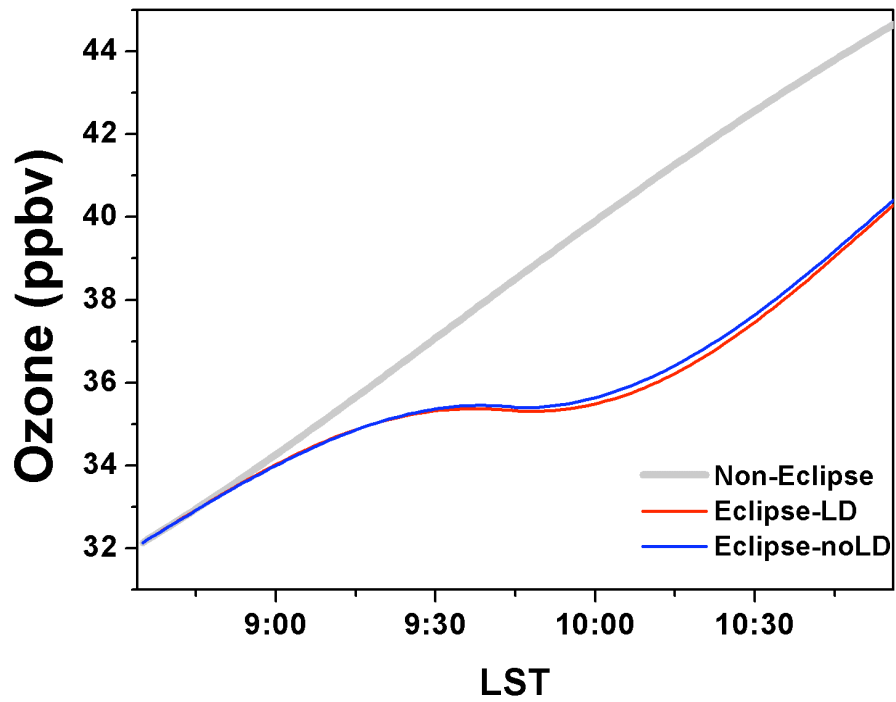

Fig. 3 Ozone concentration (ppbv) in three sensitivity experiments.

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