<u>Response to Reviewer#2</u>

We thank the reviewer for his or her helpful comments and insight. We respond to the general and specific points below. All the comments are addressed in the revised manuscript.

GENERAL COMMENTS

<u>General Comment 1:</u> This study explores the effect of clouds on heating rates driven by absorbing aerosols. They do so using observations and measurements sorted per different cloud types and coverage, separating the effects of black vs. brown carbon.
The data is collected in U9 sampling site in Milan which is a superstation that contains instruments to measure radiation, filter collecting aerosols that are analysed for their optical properties, meteorological station and a Lidar.
The topic of the paper is important. Exploring heating rates for different aerosol types under different cloud conditions will provide a very important information for aerosol effect on climate, and clouds. As the authors pointed out direct measurements of heating rates in different cloud conditions are quite uncommon.
The basic cloud classification makes sense in particularly as they added Lidar information for the clouds base. The results clearly show how cloudiness can affect heating rates and the bland between the radiation types.

<u>Answer to General Comment 1:</u> We thank the reviewer for the comment which remarks on the big effort put in this work, and the quality of both the methodological approach and of the obtained experimental results.

<u>General Comment 2:</u> One drawback of the paper is that it is very technical and not always easy to follow. Even if one understands the radiative transfer concepts, the physical assumptions and results are buried in the technicalities. It contains many technical terms that may appeal only to the instrumentation experts. Being familiar with radiation transfer concepts, I'm sure that there is a better way to describe the measurements and analyses such that a non-expert in the instrumentation could better enjoy it. The concertation of acronyms is high. It is hard to remember all of them and some that appear again later in the text force the reader to look back for their meaning and it disturbs the reading. On the other hand, some basic concepts that are key in this study are not well explained. The authors send the reader to read many other references for the basic methods and the equations. I believe that such study could be more of a standalone in which the basic physics is explained in a better way using less technical jargon.

<u>Answer to General Comment 2:</u> Thank you very much for this comment which enabled us to improve the scientific quality and presentation of the paper. Reviewer#1 asked for a shortening of the results and discussion as well as an improvement of the logical steps in the methods sections. Thus, the paper was substantially changed and improved to make it more readable and easier to follow. We have rearranged the sections to improve readability, especially the results and discussion section in the manuscript:

- In section 3.1 we introduce the environmental context of the measurement campaign and the magnitude of the observed parameters ("eBC, irradiance, HR and cloud data"). We separated the cloud analysis from the light-absorbing aerosol analysis. All the cloud analysis is presented to the reader in this section.

- The old sections 3.2 and 3.3 were re-written in agreement with the changes performed in section 3.1, and merged in a new section 3.2 with two sub-sections. The first discusses the influence of clouds in term of cloudiness while the second the influence of cloud type sub- on the total HR only (sections 3.2.1 and 3.2.2, respectively).
- Finally, the old section 3.4 (now section 3.3) was completely re-written merging and shortening the two original sub-sections 3.4.1 and 3.4.2. We discuss the role of cloudiness, cloud type and their effect on the HR apportioned with respect to BC and BrC fractions.

We have reduced the number of equations in the text. Instead of Eqs. 1-3 we now present only 2 equations in the main body of the manuscript, as follows:
"The radiative power absorbed by the aerosol in a unit volume of air (W m^{-3}; ADRE: absorptive direct radiative effect) describes the interaction between the radiation (either direct from the Sun, diffuse by atmosphere and clouds, or reflected from the ground) and the LAA (BC and BrC, Ferrero et al., 2018) and is determined as follows:

$$ADRE = \sum_{dir,dif,ref} \int_{\theta} \int_{\lambda} \frac{F_{dir,dif,ref(\lambda,\theta)}}{\cos(\theta)} b_{abs(\lambda)} d\lambda d\theta \quad \text{(A1)}$$

where the subscript *dir*, *dif* and *ref* refers to the direct, diffuse and reflected component of the spectral irradiance $F(\lambda, \theta)$ of wavelength λ that strikes (from any azimuth) with an angle θ (from the zenith) the aerosol layer and $b_{abs(\lambda)}$ is the wavelength dependent aerosol absorption coefficient. Please see Supplemental Material for further details.
To obtain the heating rate of the light-absorbing aerosol HR we divide ADRE by the air density (ρ, kg m^{-3}) and the isobaric specific heat of dry air (C_p, 1005 J kg^{-1} K^{-1}):

$$HR = \frac{1}{\rho C_p} \cdot ADRE \quad \text{(A2)}$$

We introduce the indices *dir*, *dif*, *ref* to avoid the readers' confusion about the original "n" symbol which refered to each of the different kinds of impinging radiation.
In keeping with your suggestion, we removed many acronyms and technical terms whenever possible. In agreement with a suggestion from reviewer#1, we prepared a list of acronyms and symbols (used in the manuscript) which was added in the new section Appendix A at the end of the paper.

In line with the suggestion to reference more papers, we added in the Supplemental Material an alternative notation of equations 1 as follows:

$$ADRE = \int_{\lambda} AF(\lambda) b_{abs(\lambda)} d\lambda \quad \text{(A3)}$$

where AF(λ) represents the actinic flux, that is the total spectral flux of photons per unit area and wavelength interval illuminating the molecules/aerosol at a particular point in the atmosphere where the term *actinic* refers to radiation capable of causing photochemical reactions or capable of being absorbed. The actinic flux (actually a flux density) consists of three components: direct solar radiation, diffuse radiation originating from scattering in the atmosphere, and diffuse radiation originating from reflection at the earth's ground surface. Accordingly, the actinic flux at a particular point in the atmosphere is calculated by integrating the spectral radiance over all directions in space. The actinic flux must be distinguished from the spectral irradiance, which is the hemispherically integrated radiance weighted by the cosine of the angle of incidence, and represents the photon flux per unit area through a plane surface. A more exhaustive description can be found in Liou (2007), Tian et al. (2020) and Gao et al. (2008). We added these references to the method section 2.2 of the manuscript and deepened the topic in the Supplemental material.

SPECIFIC COMMENTS

Specific Comment 1 (SC1): The aerosols that are collected at the station level serve as the only aerosol measurement and the basic assumption is that the filters collected at the station represent the whole boundary layer and therefore the heating rate is uniform for the layer below the clouds. I wonder how general this assumption is? This is always a key question of any work that try to link measurements near the surface to the atmospheric column. Is it always well mixed? Can the authors show that there is no dependency on the time of the day or the winds or the meteorology in general? Is it true for all seasons? For all cloud types? Moreover, if they have Lidar there can't they validate this assumption using the Lidar information. It would be nice to see uniform backscatter below the clouds to strengthen this basic assumption. The radiation measurements are collected in the station and are product of electromagnetic radiation interaction with the whole atmospheric column. What about the contribution of aerosols above the boundary layer. Is it assumed to be canceled by the proposed method? Or is it assumed to be negligible? If not, how such aerosols can affect the results?

Answer to Specific Comment 1 (SC1): Thank for all your questions. They are related to the methodology. In order to properly answer them it is necessary to address the following points: 1) the advantages and limitations of the applied methodology (relating to the measurements and derivation of the heating rate HR) and 2) the environmental context of the measuring site in the Po Valley (addressing the representativeness).

Methodology advantages and limitation

The most important advantages/limitations of the new method are resumed here. The first consideration is that the ADRE (and thus the HR) is the vertical derivative of the aerosol direct radiative effect (ADRE=dDRE/dz; see Ferrero et al. (2018)); we provide a detailed analysis at the end of the answer (*Methodology details and demonstration*). Thus, both the ADRE and the HR become independent from the thickness (Δz) of the investigated atmospheric layer as happens for routine atmospheric pollution measurements (i.e. BC, PM and particle number concentrations). The most important *advantages* in terms of HR measurements are:

- no radiative transfer assumptions are needed (i.e. clear sky situation), the input parameters into equations A1 and A2 are all measured,
- measurements of the spectral irradiance and the absorption coefficient are carried out at high time resolution, allowing to follow the HR dynamic with same temporal resolution,

- measurements of the spectral irradiance, the absorption coefficient and thus the HR are carried out in any sky conditions, enabling to investigate the impact by the cloud layers on the near-surface HR.

The most important *limitation* is the following:

- as both the ADRE and the HR are independent of the thickness (Δz) of the investigated atmospheric layer, they refer to the vertical location of the atmospheric layer in which both the ADRE and the HR are experimentally determined. In the present work, they are determined in the near-surface atmospheric layer.

It is noteworthy to consider the advantages that the new method allows to obtain: experimental measurement (not estimations) of ADRE and HR continuous in time with a high time resolution as a function of sources, species of light absorbing aerosol, and cloud cover. The use of the vertical derivative of the Direct Radiative Effect allows us to obtain a temporal continuity of ADRE and HR but "paying" it with the loss of vertical information.

Due to your question, we first clarified these points in the methodological section 2.2 expanding the sentence (lines 245-247 in the submitted version of the manuscript):

"As already pointed out in Ferrero et al. (2018), it is worth recalling that in the present method (equation 1), both the ADRE and the HR are independent of the investigated atmospheric aerosol layer thickness."

and at lines 250-254:

"The main advantage of the new method to quantify the impact of clouds on the light-absorbing aerosol HR is that it allows to obtain experimental measurement (not estimations) of ADRE and HR, which are continuous in time with a high time resolution, and resolved in terms of sources, species of light-absorbing aerosol, cloud cover, and cloud types."

Environmental context of HR measurements

In this section we address the representativeness of the HR determination at ground and answer the Reviewer's questions. As reported in the submitted version of the manuscript at lines 247-250: "BC and HR vertical profiles data previously collected both at the same site and in other basin valley sited (Ferrero et al., 2014) revealed that ADRE and HR were constant inside the mixing layer. The methodology is therefore believed to be valid for applications in atmospheric layers below clouds, assuming that near-surface measurements are representative of the whole mixing layer." This assumption is the core of your question.

The aim of the paper is the investigation of the impact of cloudiness and cloud-type on the HR induced by light absorbing aerosol. Ground-based highly time-resolved HR data are suitable to reach this goal – we need to introduce the representativeness shown in Ferrero et al. (2014) over Milan.

We performed combined in-situ and remote vertical profile measurements in Milan with tethered balloons and a lidar (in cooperation with the ISAC-CNR of Rome) since 2005. The collected data shows a homogeneous distribution of aerosol concentration within the mixing layer. Figure A1 reports averaged wintertime balloon profiles (PM concentrations and extinction coefficient) and lidar range corrected signal for Milan (Ferrero et al., 2019).

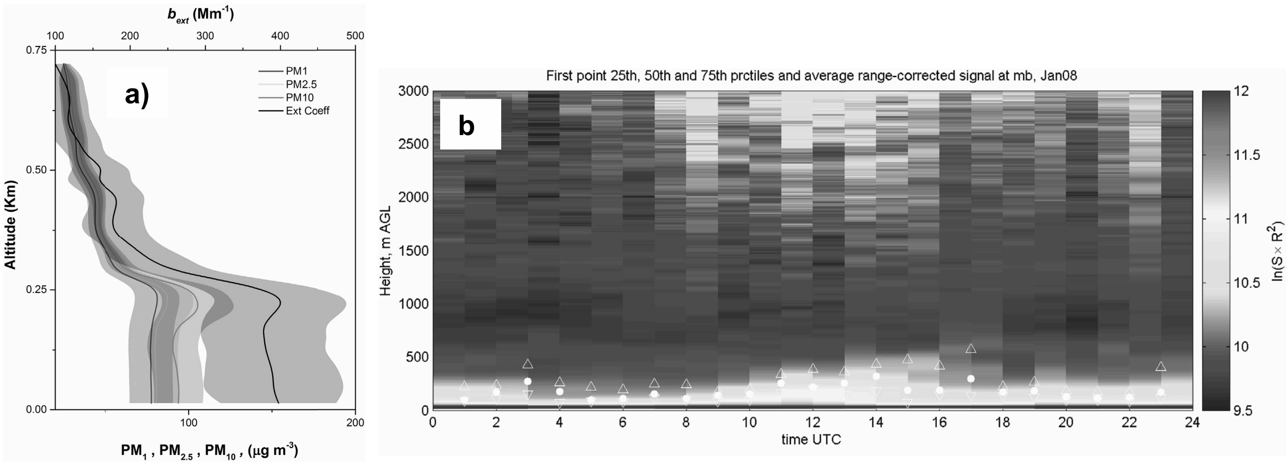

Figure A1. Milan averaged wintertime a) balloon profiles of PM_1, $PM_{2.5}$ and PM_{10} and extinction coefficient b) lidar range corrected signal. Data for the present figure are from Ferrero et al. (2019).

The same condition was verified by the lidar-ceilometer data collected during the present campaign (Figure A2, here below).

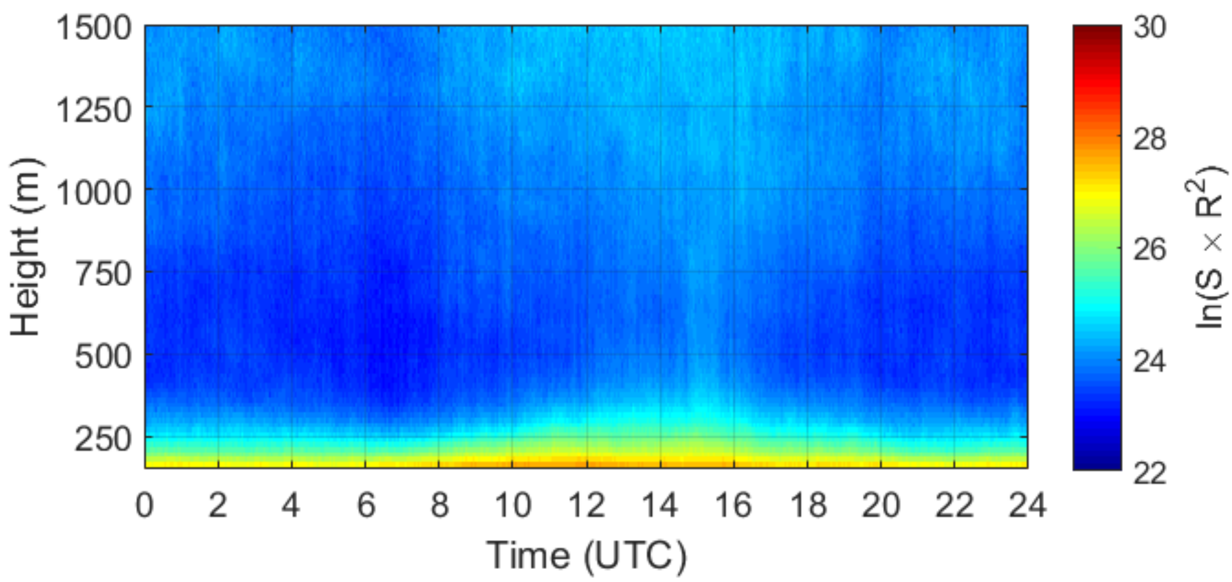

Figure A2. Milan averaged wintertime lidar range corrected signal (SxR^2) during the campaign presented in the manuscript.

Vertical profiles data reported in Figure A1 and A2 experimentally verify the assumption "that near-surface measurements are representative of the whole mixing layer" in wintertime in Milan.
Figures A1b and A2 show a typical mixing layer height diurnal behavior in wintertime conditions, with the mixing layer height not exceeding 500 m above ground. The same was previously retrieved from the vertical gradient of tethered balloon aerosol profiles (Ferrero et al., 2010; Figure A3). Within

the mixing layer, aerosol concentrations were uniform (as reported in Figure A1) along each time of the day.

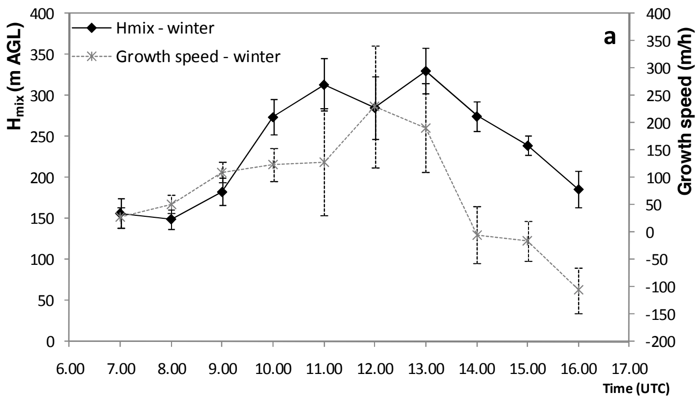

Figure A3. Diurnal variation of the mixing layer height. Plot taken from Figure 4, Ferrero et al. (2010).

Finally, in Ferrero et al. (2014), we explored the vertical behavior of the light absorbing aerosol HR. It is reported here below in Figure A4.

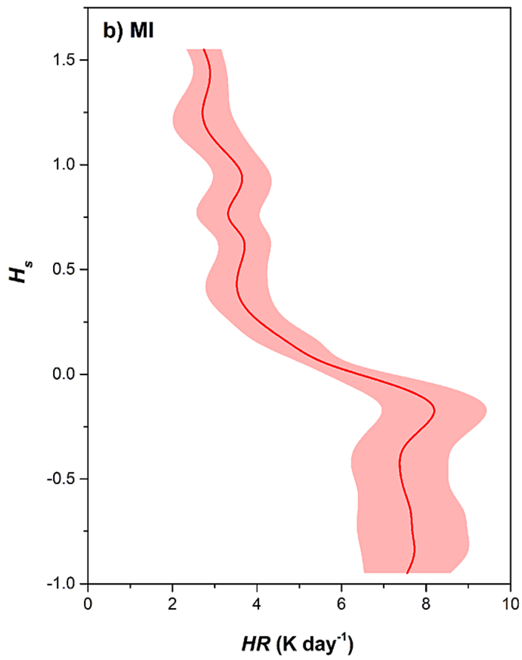

Figure A4. Heating rate (HR) vertical profile, with the normalized height H_s = -1 at ground level and 0 at the top of the mixing layer. Plot taken from Figure 10, Ferrero et al. (2014).

Figure A4 shows that the HR can be considered constant inside the mixing layer, making near-surface measurements representative of the mixing layer height.

Finally, as shown by both Figure A1 and A2 and as reported in Ferrero et al. (2019) the collected wintertime vertical data in Milan showed that 87.0% of AOD (aerosol optical depth) signal was contributed to within the mixing layer, 8.2% in the residual layer and 4.9% in the free troposphere. The impact of clouds on the incoming radiation reaching the mixing layer is therefore dominant.

We added Figure A2 in Supplemental Material and the aforementioned consideration in section 2.2. Here below, as written at the beginning of this answer, the method details and demonstration.

Methodology details and demonstration (Ferrero et al., 2018)

We start from the radiative transfer concept of the instantaneous aerosol Direct Radiative Effect (DRE; W m^{-2}) which can be quantified as the change in the net radiative flux between the atmospheric conditions with aerosols (*aer*) and without the aerosols ($Q_{aer}(z)$ and $Q_0(z)$, respectively) in the atmosphere across the surface at altitude z:

$$DRE(z) = Q_{aer}(z) - Q_0(z) \tag{A4}$$

Considering an atmospheric layer of thickness Δz, the difference between the *DRE* at the top and the bottom of this atmospheric layer represents the instantaneous radiative power density absorbed by the aerosol (ΔDRE; W m^{-2}):

$$\Delta DRE = DRE(z + \Delta z) - DRE(z) \tag{A5}$$

From ΔDRE, the instantaneous heating rate (*HR*; K day^{-1}) of the same atmospheric layer can be computed as follows:

$$HR = \frac{\partial T}{\partial t} = -\frac{g}{C_p}\frac{\Delta DRE}{\Delta p} \tag{A6}$$

where $\partial T/\partial t$ represents the instantaneous *HR*, g is the gravitational acceleration constant, C_p (1005 J kg^{-1} K^{-1}) is the isobaric specific heat of dry air, Δp is the pressure difference between the top and the bottom of the considered layer.

A more useful definition of the *HR* is based on the thickness of the atmospheric layer (Δz), and can be obtained introducing the hydrostatic equation ($dp = -\rho g dz$) into Eq. A6:

$$HR = \frac{\partial T}{\partial t} = \frac{1}{\rho C_p}\frac{\Delta DRE}{\Delta z} \tag{A7}$$

where ρ represents the air density (kg m^{-3}). The last term of Eq. A7 ($\Delta DRE/\Delta z$) represents the radiative power absorbed by the aerosol for unit volume of the atmosphere (W m^{-3}) and is defined as the absorptive direct radiative effect (*ADRE*) of light-absorbng aerosols. The *ADRE* is the vertical spatial derivative of the *DRE* (*dDRE/dz*). Hence, the *HR* becomes:

$$HR = \frac{1}{\rho C_p} \cdot ADRE \tag{A8}$$

Thus, any method able to determine the *ADRE* at high time resolution will produce continuous highly time-resolved time series of *HR*.

Let us consider a near-surface atmospheric layer of thickness Δz on which direct or diffuse or reflected monochromatic radiation ray $F_n(\lambda,\theta)$ of wavelength λ strikes with a zenith angle θ. We use the subscript n to denote the type of radiation: direct, diffuse or reflected. The amount of radiation absorbed by the aerosol within the present layer is as follows:

$$\Delta DRE_n(\lambda,\theta) = F_n(\lambda,\theta)(1-\omega(\lambda))\left(1-e^{-\tau(\lambda)/\cos\theta}\right) \tag{A9}$$

where (1- $\omega(\lambda)$)(1-$e^{-\tau(\lambda)/\cos\theta}$) represents the fraction of light absorbed within the layer and is function of: $\omega(\lambda)$ – the single scattering albedo of the aerosol within the atmospheric layer, and $\tau(\lambda)$ – the aerosol optical depth. The $\omega(\lambda)$ and $\tau(\lambda)$ terms can be computed from the aerosol extinction, scattering and absorption coefficients ($b_{ext}(\lambda)$, $b_{sca}(\lambda)$ and $b_{abs}(\lambda)$):

$$\omega(\lambda) = \frac{b_{sca}(\lambda)}{b_{sca}(\lambda)+b_{abs}(\lambda)} \tag{A10}$$

$$\tau(\lambda) = \int_o^{\Delta z} b_{ext}(\lambda)\ dz \tag{A11}$$

Now, if the atmospheric layer is thin enough so that $\tau(\lambda) \ll 1$, the term (1-$e^{-\tau(\lambda)/\cos\theta}$) can be simplified introducing the Taylor series and the radiative power $\Delta DRE_{n(\lambda,\theta)}$ absorbed by the aerosol within that atmospheric layer can be computed from eq. A6 as follows:

$$\Delta DRE_n(\lambda,\theta) = F_n(\lambda,\theta)(1-\omega(\lambda))\ \frac{\tau(\lambda,\theta)}{\cos\theta} \tag{A12}$$

In this form, $\Delta DRE_n(\lambda,\theta)$ is not useful because it is a columnar quantity which again depends on $\tau(\lambda)$ that is integrated along the vertical direction.

Considering again an atmospheric layer thin enough so that $\tau(\lambda) \ll 1$ it is also possible to assume $F_n(\lambda,\theta) \approx$const and $\omega(\lambda) \approx$const through the whole Δz; thus, recalling the *ADRE* definition (*ADRE=dDRE/dz*), and combining Eq. A10 with Eq. A11 and Eq. A12, it is possible now to write:

$$ADRE_n(\lambda,\theta) = \frac{dDRE_n(\lambda,\theta)}{dz} = F_n(\lambda,\theta)\frac{(1-\omega(\lambda))}{\cos\theta}\frac{d\tau_\lambda(\lambda)}{dz} = \frac{F_n(\lambda,\theta)}{\cos\theta}b_{abs}(\lambda) \tag{A13}$$

Equation A13 offers the opportunity to determine the *ADRE*, and thus the *HR* (eq. A5), just combining the absorption coefficient of light absorbing aerosols $b_{abs}(\lambda)$ and radiation measurements $F_n(\lambda,\theta)$. Thus, the resulting *ADRE* and *HR* are only related to the light absorbing aerosols (and not to gases). Obviously, the atmospheric absorption and related *HR* can be obtained integrating Eq. A13 over the whole ensemble of shortwave wavelengths and incident angles:

$$ADRE_n = \int_\theta \int_\lambda \frac{F_n(\lambda,\theta)}{\cos\theta} b_{abs}(\lambda) d\lambda\, d\theta \tag{A14}$$

The shortwave radiation that can cross the atmospheric layer can be divided in three components, namely: the solar direct radiation ($F_{dir}(\lambda,\theta)$); the diffuse radiation from scattering on gases, aerosol and clouds in the sky ($F_{dif(}(\lambda,\theta)$); and the radiation reflected backward from the ground ($F_{ref}(\lambda,\theta)$).

Equation A14 can be solved for all the three components allowing to determine both the total *ADRE* and its components ($ADRE_{dir}$, $ADRE_{dif}$ and $ADRE_{ref}$) as follows:

$$ADRE = ADRE_{dir} + ADRE_{dif} + ADRE_{ref} \tag{A15}$$

Using Eq. A8 the same is valid for the *HR*:

$$HR = HR_{dir} + HR_{dif} + HR_{ref} \tag{A16}$$

so, the final equation for the *HR* can be written as follows:

$$HR = \frac{1}{\rho C_p} \cdot \sum_{n=1}^{3} \int_\theta \int_\lambda \frac{F_n(\lambda,\theta)}{\mu} b_{abs}(\lambda) d\lambda\, d\theta \tag{A17}$$

where n represents direct or diffuse or reflected radiation.

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