

# 1 ON THE ENERGY POTENTIAL OF DAYTIME RADIATIVE

## 2 COOLING FOR URBAN HEAT ISLAND MITIGATION

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### 15 KEYWORDS

16 Daytime radiative cooling; sensitivity analysis; spectrally selective materials; cooling  
17 potential; Urban Heat Island.

### 18 ABSTRACT

19 The objective of this paper is to present the potential of daytime radiative cooling  
20 materials as a strategy to mitigate the Urban Heat Island effect. To evaluate the cooling  
21 potential of daytime radiative cooling materials, 15 theoretical materials and seven  
22 existing materials were simulated: two radiative cooling materials, a coolmaterial, two  
23 white paints, a thermochromic paint and a construction material. The novelty of this

24 study is that it shows that the optimal spectral characteristics of radiative cooling  
25 materials depending on the climate conditions and the type of application. A sensitivity  
26 analysis was performed to evaluate the impact of each wavelength emissivity on the  
27 ability to achieve sub-ambient radiative cooling. The sensitivity analysis comprised a  
28 total of 90 theoretical materials with 15 different wavelength combinations and 6  
29 emissivity values. The heat transfer model, which includes conduction, convection, and  
30 radiation, was developed using a spectrally-selective sky model. Two conditions were  
31 considered: a very conductive surface and a highly insulated one. All the materials  
32 were simulated in two cities that suffer from the Urban Heat Island effect—Phoenix and  
33 Sydney. The mean surface temperature reduction achieved was 5.30 °C in Phoenix  
34 and 4.21 °C in Sydney. The results presented suggest that the type of application  
35 (active or passive) is a determinant factor in the design of radiative cooling materials.  
36 Modifying the spectra of the materials led to a substantial change in the cooling  
37 potential. A material that performs well in a dry climate as a passive solution could  
38 perform poorly as an active solution.

## 39 **HIGHLIGHTS**

- 40 • Daytime radiative cooling materials are studied in two cities with Urban Heat  
41 Island
- 42 • Contribution of wavelength band to the ability to achieve sub-ambient  
43 temperatures
- 44 • Materials studied under highly-insulated and highly-conductive conditions
- 45 • Material's behavior strongly depends on the location and the type of application

Nomenclature	
$c$	Fraction of sky covered by clouds
$E_b$	Blackbody radiation $W/m^2$
$G$	Irradiance $W/m^2$
$h$	Convective heat transfer $W/(m^2K)$
$I$	Solar irradiation $W/m^2$
$J$	Radiosity $W/m^2$
$q$	Heat flux $W/m^2$
$T$	Temperature $K$
$\nu$	Wavenumber $1/m$
Greek letters	
$\varepsilon$	Emissivity
Subscripts	
$cd$	Conduction
$cv$	Convection
$r$	Radiation
$s$	Surface studied
$sun$	Solar
$\Delta\nu$	A wavenumber range or band

46

## 47 1. Introduction

48 Nowadays, half of the world's population lives in urban areas (*World Urbanization*  
49 *Prospects*, 2014) and consumes 75% of the primary energy sources, emitting between  
50 50 and 60% of greenhouse gases ("Energy – UN-Habitat," n.d.). Furthermore, the  
51 world's urban population is expected to increase by more than two-thirds by 2050,  
52 reaching 6.3 billion (*World Urbanization Prospects*, 2014), with nearly 90% of this rise

53 taking place in cities across Asia and Africa. CO<sub>2</sub> emissions increase proportionately  
54 with population due to energy use (O'Neill et al., 2012). A 1% increase in the urban  
55 population is estimated to increase energy consumption by 2.2% (Santamouris et al.,  
56 2001). The global energy demand is predicted to increase by more than 25% if the  
57 IEA's New Policies Scenario (rising incomes and an extra 1.7 billion people) is followed  
58 (International Energy Agency, 2018).

59 Higher urban temperatures are due to the positive thermal balance of urban areas in  
60 comparison with rural areas, caused by (i) the significant release of anthropogenic  
61 heat, (ii) the excess storage of solar radiation by city structures, (iii) the lack of green  
62 spaces and cold sinks, (iv) the non-circulation of air in urban canyons, and (v) the  
63 reduced ability of emitted infrared radiation to escape into the atmosphere (Oke et al.,  
64 1991). This phenomenon, known as the Urban Heat Island (UHI), is well documented  
65 in more than 400 cities around the world (Santamouris, 2019), and the total number of  
66 reported cities is increasing rapidly. The average UHI varies between 0.5 °C to 7 °C,  
67 where 90% of the data is below 4.5 °C (Santamouris, 2020). As ambient air  
68 temperature increases, the carrying capacity of electric power cables decreases, a  
69 phenomenon that occurs more during the summer with the increase in electricity load  
70 caused by air-conditioning usage (Bartos et al., 2016). Moreover, UHI and heatwaves  
71 have a relevant environmental and financial impact, especially on vulnerable and low-  
72 income populations (Santamouris and Kolokotsa, 2015). Additionally, exposures to  
73 high ambient temperatures represent a serious health danger (Anderson G. Brooke  
74 and Bell Michelle L., 2011).

75 The urban climate is strongly determined by morphological characteristics and the  
76 properties of the materials comprising the urban landscape (Lemonsu et al., 2015).  
77 Many strategies focusing on new material developments have been proposed to  
78 mitigate the rise in cooling demand, and the increase in urban temperatures. Increasing  
79 the global albedo of the city has resulted in a reduction in the peak ambient

80 temperature of up to 3 °C and a 20% reduction in peak cooling demand in residential  
81 buildings (Santamouris et al., 2018). Cool roofs have been widely studied for reducing  
82 the cooling demand (Bell et al., 2003; Berdahl and Bretz, 1997; Erell et al., 2006;  
83 Kolokotroni et al., 2013; Kolokotsa et al., 2018; Miller et al., 2015; Radhi et al., 2017;  
84 Santamouris, 2013; Santamouris et al., 2008). Green roofs and vegetation have been  
85 proposed as a mitigation strategy as well (Foustalieraki et al., 2017; Herrera-Gomez et  
86 al., 2017; Kolokotsa et al., 2013; Zinzi and Agnoli, 2012).

87 Recently developed radiative cooling materials have achieved daytime sub-ambient  
88 temperatures even under direct solar radiation. Radiative cooling is the physical  
89 phenomenon by which an object dissipates heat as infrared radiation. Over mid-  
90 infrared wavelengths, between 8 and 14  $\mu\text{m}$ , the Earth's atmosphere is transparent to  
91 electromagnetic radiation. Radiative cooling was applied as a nocturnal passive system  
92 for cooling in some experimental buildings and prototypes (Yellot, 1976; Yellot, John I.,  
93 1976). The results showed a limited nocturnal cooling capacity since longwave  
94 radiation from commonly-found materials can rarely achieve cooling powers of more  
95 than  $100 \text{ W}\cdot\text{m}^{-2}$ , even under ideal meteorological conditions (Erell and Etzion, 1992).  
96 Material sciences have significantly evolved since the first designs were researched in  
97 the early '70s and radiative cooling materials have recently been developed (Kou et  
98 al., 2017; Li et al., 2017; Raman et al., 2014; Rephaeli et al., 2013; Shi et al., 2018;  
99 Zhai et al., 2017; Zhu et al., 2019, 2015). Novel materials such as photonics,  
100 metasurfaces, and polymers have already achieved  $120 \text{ W}\cdot\text{m}^{-2}$  under direct sunlight  
101 (Santamouris and Feng, 2018). Besides material development, aperture dependency  
102 and geometrical designs have been studied. This kind of device was introduced by  
103 Trombe in 1967 (cited by (Smith, 2009)) by placing blackbody materials facing the sky  
104 and protecting them from the environment. Several authors have continued this line of  
105 research (Aviv and Meggers, 2017; Smith, 2009; Zhou et al., 2019a, 2019b), showing  
106 temperature drops of up to 11 °C below ambient temperature.

107 In order to achieve daytime radiative cooling, the optical properties in each wavelength  
108 of a material are determinant. The material needs to emit highly in the atmospheric  
109 transparency window (7.9-14  $\mu\text{m}$ ) and reflect at least 94% of incident sunlight (0.3-3  
110  $\mu\text{m}$ ) (Raman et al., 2014). Absorbing 10% of incident solar radiation is approximately  
111  $100 \text{ W}\cdot\text{m}^{-2}$  and therefore, the thermal equilibrium is reached at a higher temperature  
112 than the ambient temperature. Daytime radiative cooling materials have been coupled  
113 to air-conditioning (AC) systems to evacuate the excess heat to space instead of to the  
114 ambient air (Aili et al., 2019; Goldstein et al., 2017; Wang et al., 2018; Zhang et al.,  
115 2018). In (Zhao et al., 2019), the authors compared an air radiative cooling system with  
116 other materials and systems (shingle roof, attic ventilation, and coolroof). Using their  
117 proposed radiative cooling system, they achieved a reduction in the attic air  
118 temperature of 15.5-21  $^{\circ}\text{C}$ . Another system using the material developed by (Zhai et  
119 al., 2017) reduced the energy consumption and achieved savings from 26% to 46% for  
120 the modeled locations (Zhang et al., 2018).

121 Radiative cooling depends on the optical properties of the material and the thermal  
122 exchange with the surroundings. The effect of climatic parameters such as the effect of  
123 air temperature, solar radiation, and ambient radiation have recently been discussed  
124 (Feng et al., 2020). Moreover, the contribution of convection has been vastly  
125 researched (Chen et al., 2016; Cui et al., 2016; Huang and Ruan, 2017; Kou et al.,  
126 2017). Various studies have calculated the radiative cooling potential of several  
127 devices and materials in different cities (Feng et al., 2020; Vall et al., 2018), countries  
128 (Li et al., 2019), and areas of the world (Argiriou et al., 1992). Nevertheless, a more  
129 detailed study showing the impact of the optical properties of each wavelength band on  
130 the ability to achieve sub-ambient cooling has not yet been presented.

131 This research aims to study the impact of the different spectral selectivity  
132 configurations in the cooling potential of radiative materials by conducting a sensitivity  
133 analysis. The effects of each wavelength's band emissivity on the ability to achieve

134 sub-ambient cooling was determined. The authors compared the performance of  
135 several theoretical radiative cooling materials with newly developed ones and typical  
136 construction materials. The materials were studied under two conditions to assimilate a  
137 passive and active solution (for future integration in AC systems). Firstly, the passive  
138 solution was designed as a highly insulated surface on one side (an almost adiabatic  
139 condition). Secondly, the active condition was assimilated to a very conductive surface.  
140 Besides, several convective values were simulated to determine the maximum sub-  
141 ambient cooling. As a result, considerations for choosing the appropriate spectral  
142 emissivity configuration are given for each location. The restrictions to achieving  
143 daytime radiative cooling are detailed for both conditions.

144 The main novelty of this study is that it shows that the desired spectral emissivity  
145 characteristics of radiative cooling materials depend on the climate conditions and the  
146 type of application. It was discovered that the best spectral characteristics are different  
147 for a dry or humid climate and if the application is for a passive system or an active  
148 one. In-depth study of these two aspects is required in future research to establish the  
149 level of importance of these two observations using broader statistical data.

## 150 **2. Methodology**

151 The research methodology described was followed to determine the impact of the  
152 spectral emissivity configuration on the possibility of achieving sub-ambient  
153 temperatures. First, a heat transfer model was developed. This model simulated a  
154 horizontal flat plate, in which the conductive heat transfer was calculated using the  
155 finite difference method (implicit method). The boundary conditions on the lower side  
156 were convection and the temperature of a fluid. For a highly insulated condition, a  
157 nearly zero value is defined for the convective heat transfer coefficient. For the upper  
158 side, the boundary conditions defined were convection with air, incident solar radiation,  
159 and radiation exchange with the atmosphere. The optical properties varied spectrally,  
160 and the model considered this variation for both solar radiation and atmospheric

161 radiation. Atmospheric radiation is based on the spectrally selective sky model  
162 presented by Berger and Bathiebo (Berger and Bathiebo, 1989), where the sky  
163 conditions were defined as clear sky, completely covered sky, and partially covered  
164 sky.

165 The model can perform transient and steady-state simulations with time steps specified  
166 at the beginning of each simulation (for this study, a 1-minute time step was used in all  
167 cases); the summary results presented are hourly, however. The main variables  
168 obtained in the results are surface temperatures and heat transfer on the surface. The  
169 heat transfer is discretized for each transfer mechanism: conduction, convection,  
170 radiation from the atmosphere, and solar radiation absorption. The power at the end of  
171 the hour is given for each heat transfer mechanism. Moreover, the cumulative energy  
172 transferred during the hour is presented. The previous results are used to obtain other  
173 related parameters, such as the hourly difference between air temperature and surface  
174 temperature, the daily mean of this difference, daily mean temperatures, and  
175 cumulative daily heat transfer.

176 The model was validated using outdoor experimental data from two newly developed  
177 radiative cooling materials (Raman et al., 2014; Zhai et al., 2017). Following the  
178 validation, the sensitivity analysis was conducted with 90 theoretical materials and  
179 seven existing materials in two locations: Sydney, with a mild climate and Phoenix, with  
180 an arid climate (Cfa and Bsh respectively according to (Kottek et al., 2006)). Moreover,  
181 the two boundary conditions—a high insulated and a very conductive surface—were  
182 simulated. Finally, the results are compared according to their radiative cooling  
183 potential and the surface temperature they reached. The suitability, restrictions, and  
184 limitations for each are presented for each location and boundary condition.

## 185 **2.1. Heat transfer model**



186 The objective of the heat transfer model is to simulate the thermal behavior of a  
 187 horizontal surface that exchanges heat with its surroundings by convection and  
 188 radiation. Radiation is divided into two components: solar radiation and radiation with  
 189 the surroundings. The surrounding radiation will be assumed to be only from the sky.  
 190 The heat transfer model calculated the heat transferred by conduction from the surface  
 191 down with the finite difference method. The emphasis of the model is on the two  
 192 radiation components, since it considers the spectral characteristics of the surface, sky  
 193 and solar radiation; therefore, only this component will be explained.

194 The radiosity of a surface with a view factor equal to one with the sky can be  
 195 represented by:

$$J_s(\nu) = \varepsilon_s(\nu)E_b(\nu, T_s) + (1 - \varepsilon_s(\nu))G_s(\nu) \quad (1)$$

196 Where  $J_s(\nu)$  is the radiosity of the surface for the wavenumber  $\nu$ ,  $\varepsilon_s(\nu)$  the emissivity of  
 197 the surface for the wavenumber  $\nu$ ,  $E_b(\nu, T_s)$  the blackbody radiation in the wavenumber  
 198  $\nu$  when its temperature is  $T_s$ ,  $G_s(\nu)$  the irradiance received by the surface at a  
 199 wavenumber  $\nu$ .

200 As the sky is the only radiation emitter that the surface faces, the irradiance received  
 201 by the surface is:

$$G_s(\nu) = \varepsilon_{sky}(\nu)E_b(\nu, T_{air}) \quad (2)$$

202 Therefore:

$$J_s(\nu) = \varepsilon_s(\nu)E_b(\nu, T_s) + (1 - \varepsilon_s(\nu))\varepsilon_{sky}(\nu)E_b(\nu, T_{air}) \quad (3)$$

203 On the other hand, the radiation heat flux in the surface for the wavenumber  $\nu$  is:

$$q_{r,s,\nu} = J_s(\nu) - G_s(\nu) \quad (4)$$

$$q_{r,s,\nu} = \varepsilon_s(\nu) \left( E_b(\nu, T_s) - \varepsilon_{sky}(\nu)E_b(\nu, T_{air}) \right) \quad (5)$$

204 For the wavelength bands between wavenumbers between  $\nu_i$  and  $\nu_j$  the heat flux is:

$$q_{r,s,\Delta v} = \int_{v_i}^{v_j} \varepsilon_s(v) \left( E_b(v, T_s) - \varepsilon_{sky}(v) E_b(v, T_{air}) \right) dv \quad (6)$$

$$q_{r,s,\Delta v} = \int_{v_i}^{v_j} \varepsilon_s(v) E_b(v, T_s) dv - \int_{v_i}^{v_j} \varepsilon_s(v) \varepsilon_{sky}(v) E_b(v, T_{air}) dv \quad (7)$$

205 Selecting a range or band  $\Delta v$  where both  $\varepsilon_s(v)$  and  $\varepsilon_{sky}(v)$  can be considered  
 206 approximately constant yields the following expression:

$$q_{r,s,\Delta v} = \varepsilon_{s,\Delta v} \left[ \int_{v_i}^{v_j} E_b(v, T_s) dv - \varepsilon_{sky,\Delta v} \int_{v_i}^{v_j} E_b(v, T_{air}) dv \right] \quad (8)$$

207 The previous equation makes it possible to calculate the heat flux exchanged by  
 208 radiation with the sky for each band in which the emissivity of the sky and the surface  
 209 can be considered constant. The total heat flux is therefore the summation of the heat  
 210 flux of each band:

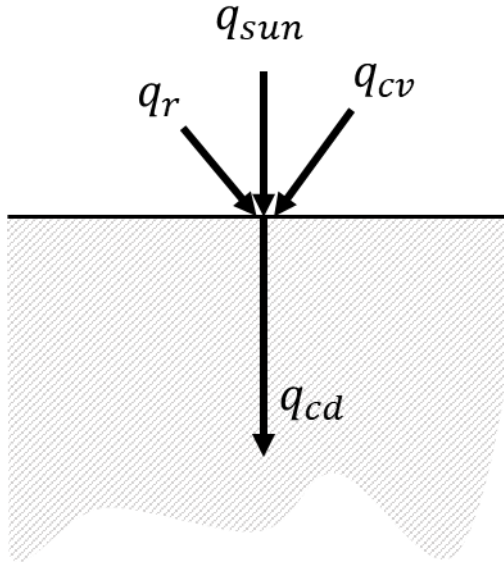
$$q_r = \sum_{\Delta v_i} q_{r,s,\Delta v_i} \quad (9)$$

211 The second component of radiation is the solar radiation. In this case the heat can be  
 212 calculated with:

$$q_{sun} = \int_0^{\infty} \varepsilon_s(v) I(v) dv \quad (10)$$

213 And similar to the previous considerations, if there are bands where emissivity can be  
 214 considered as constant, the previous integral can be approximated by a summation:

$$q_{sun} \approx \sum_{\Delta v_i} \varepsilon_{s,\Delta v_i} I_{\Delta v_i} \quad (11)$$



215

216 *Figure 1. Heat flux in the surface*

217 Finally, the balance of heat flux at the surface, following the schematic representation  
 218 of Figure 1, can be expressed as:

$$q_{cd} = q_r + q_{sun} + q_{cv} \quad (12)$$

219 Where:

$$q_{cv} = h_{cv}(T_{air} - T_s) \quad (13)$$

220 And  $q_{cd}$ , as stated above, is calculated using the finite difference method.

### 221 **2.1.1. Sky model review and development**

222 Calculating the heat transfer with the atmosphere using equation 8 requires the  
 223 spectral emissivity of the sky ( $\varepsilon_{sky,\Delta v}$ ) to be known.

224 Although there is a vast literature on sky models (30 evaluated in (Vall and Castell,  
 225 2017), 35 in (Algarni and Nutter, 2015) and 70 in (Antonanzas-Torres et al., 2019))  
 226 both for clear sky conditions and cloudy conditions, most of them refer to global  
 227 emissivity. Clouds act as a barrier to heat transfer, inhibiting the outgoing radiation  
 228 through the atmospheric band and augmenting the effective temperature of the sky due  
 229 to the absorption of heat by water vapor (Berdahl and Fromberg, 1982). Opaque clouds

230 can be considered blackbody emitters at the temperature of the cloud base (Bliss,  
231 1961) and their radiative effect close to the transparency window. The influence of  
232 cloud radiation decreases with altitude; higher clouds are colder than lower clouds  
233 (Sugita and Brutsaert, 1993). Nevertheless, measurements of the downward  
234 component emitted by clouds and aerosols in the atmosphere are scarce and not well  
235 understood (J Herrero and Polo, 2012).

236 Cloudy sky models are based on daytime clearness indexes and are transposed as a  
237 single value for the night; moving clouds cannot be considered (Eicker and Dalibard,  
238 2011). Moreover, at night there are no cloud coverage estimations. Malek (Malek,  
239 1997) proposes a method to evaluate sky cloud cover without having to relate to any  
240 empirical and local constants. It is based on the cloud base height, cloud base  
241 temperature, and percentage of the sky covered by clouds. Emissivity value estimates  
242 are 1 under cloudy conditions (Martin and Berdahl, 1984) and above 0.95 for covered  
243 skies (J. Herrero and Polo, 2012) . According to (Monteith and Unsworth, 2013), for a  
244 completely overcast sky (cloud fraction  $c = 1$ ) in Oxford, England, the apparent  
245 emissivity of the sky can be calculated knowing the emissivity value for clear sky by:

$$\varepsilon_{sky,cloud} = \varepsilon_{sky}(1) = 0.84 + 0.16\varepsilon_{sky,clear} \quad (14)$$

246 And for a sky covered with a fraction “ $c$ ” of cloud, emissivity can be calculated by  
247 interpolation:

$$\varepsilon_{sky}(c) = c\varepsilon_{sky}(1) + (1 - c)\varepsilon_{sky,clear} \quad (15)$$

#### 248 **Clear sky emissivity model**

249 This research uses the Berger and Bathiebo 1989 (Berger and Bathiebo, 1989)  
250 spectral sky model to calculate the spectral radiation of the atmosphere. The model  
251 calculates the spectral and global emissivity of the sky in 21 wavelength ranges, using  
252 Equation 16

$$\varepsilon_{sky,\Delta v_i} = 1 - \exp(-k_{\Delta v_i} w_j) \quad (16)$$

253

254 Where  $\varepsilon_{sky,\Delta v_i}$  is the clear sky emissivity for the range of wavenumbers  $\Delta v_i$ ;  $k_{\Delta v_i}$  is an  
 255 absorption coefficient, and  $w_j$  an equivalent absorber that must be calculated using  
 256 different correlations defined by (Berger and Bathiebo, 1989) for each band.

### 257 **Completely covered sky emissivity**

258 To the best knowledge of the authors, there are no correlations to calculate the spectral  
 259 emissivity under completely covered skies with a similar degree of discretization that  
 260 has been presented for clear skies. The model developed can calculate the emissivity  
 261 of the sky using a correlation of Equation 14 or a constant value. By default, a value of  
 262 0.95 (J. Herrero and Polo, 2012) was used for the results calculated in this paper.

### 263 **Partially covered sky emissivity**

264 As shown in Equation 15, a linear relationship between the emissivity of clear skies and  
 265 completely covered skies is presented in (Monteith and Unsworth, 2013). This principle  
 266 can be applied for each spectral band. The parameter “c” is used as the fraction of sky  
 267 covered by clouds, 1 being a completely covered sky. For a sky covered with a fraction  
 268 c of cloud, interpolation gives:

$$\varepsilon_{sky,\Delta v_i}(c) = c\varepsilon_{sky}(1) + (1 - c)\varepsilon_{sky,\Delta v_i} \quad (17)$$

269 The sky emissivity for a partially covered sky is the weighted average between the  
 270 emissivity of clear sky and the completely covered sky, the ratio of the covered sky  
 271 being the weighting factor. Note that emissivity for a completely covered sky  $\varepsilon_{sky}(1)$  is  
 272 not dependent on the wavenumber since there is no information about its spectral  
 273 variation.

## 274 **2.2. Model validation**

275 The model was validated using data from two recently developed materials in the  
 276 literature (Raman et al., 2014; Zhai et al., 2017); the authors reported very high cooling  
 277 rates even when exposed to the sun and achieved a substantial temperature drop from  
 278 the ambient temperature. The model agreed well with experimental data (Figure 2 and  
 279 3) and is considered valid.

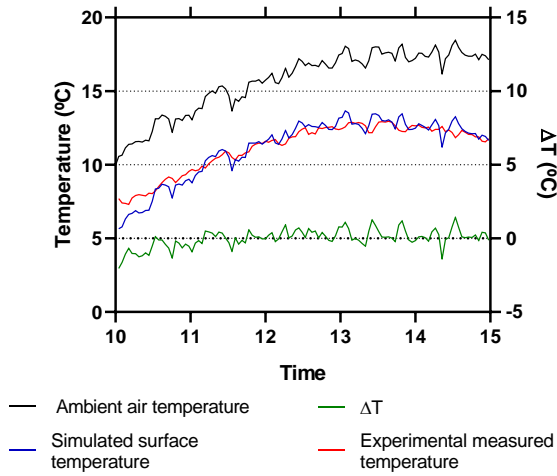
280 *Table 1: Comparison of the two radiative cooling materials under experimental conditions.*

	<b>Skycool (Raman et al., 2014)</b>	<b>Radicool (Zhai et al., 2017)</b>
<b>Solar Reflectivity</b>	0.90	0.90
<b>Emissivity in the transparency window</b>	0.80	0.93
<b>Reported sub-ambient temperature</b>	4.9 °C	
<b>Cooling potential</b>	40.1 W/m <sup>2</sup>	93 W/m <sup>2</sup>
<b>Location of experiment</b>	Stanford, CA, USA	Cave Creek, AZ, USA
<b>Köppen climate exp</b>	Csb	BSh
<b>Dates of the experiment</b>	Clear winter day	16 <sup>th</sup> Oct. to 19 <sup>th</sup> Oct (Fall)

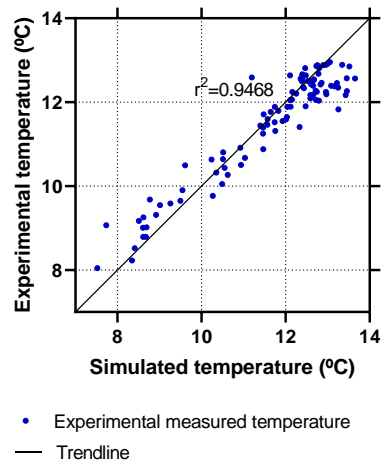
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282 In the experiment conducted in 2014, the authors exposed the radiative cooling  
 283 material protected by a low-density polyethylene cover to the sky. These polyethylene  
 284 covers acted as a convection barrier (See Supplementary Material 1: Model Validation  
 285 Procedure).

(a) Simulated and experimental surface temperature RC1



b) Correlation between simulated and experimental surface temperature



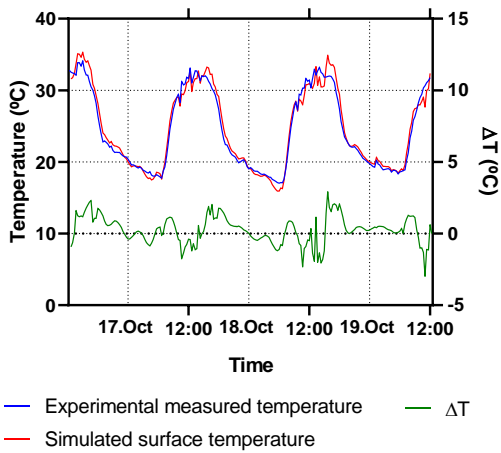
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287 *Figure 2. Validation of the thermal model with material 1 (RC1) “Skycool” (Raman et al., 2014).*

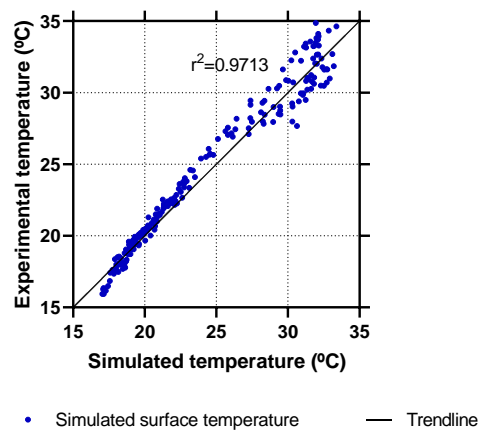
288 The second experimental setting was different; they eliminated convection by applying

289 a constant heat supply to the material to achieve ambient temperature.

(a) Simulated and experimental surface temperature RC2



b) Correlation between simulated and experimental surface temperature



290

291 *Figure 3. Validation of the thermal model with material 1 (RC2) “Radicool” (Zhai et al., 2017).*

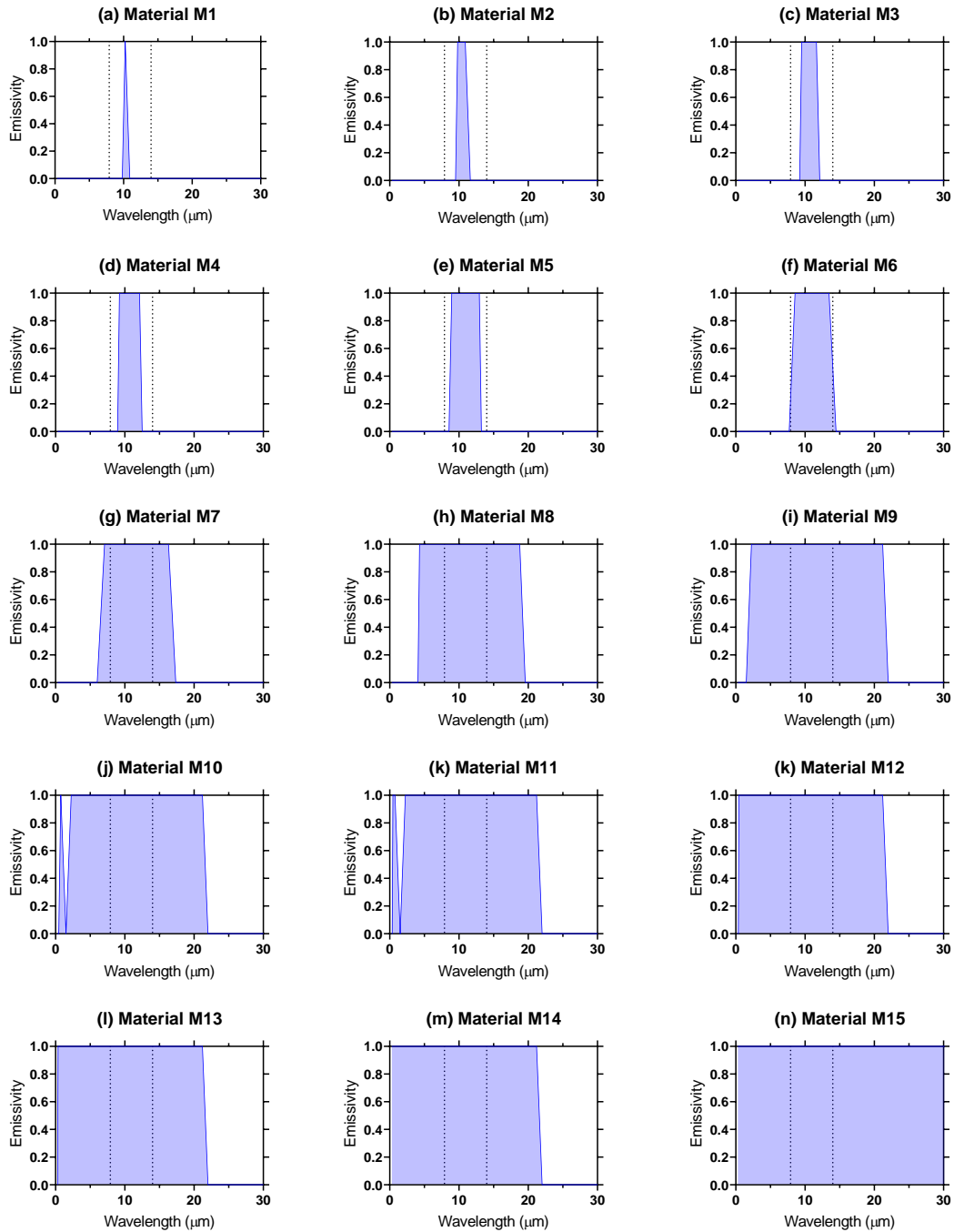
292 **2.3. Sensitivity analysis**

293 The objective of this sensitivity analysis was to determine the impact of wavelength  
294 emissivity on the ability to achieve daytime radiative cooling. The radiation spectrum  
295 was divided into 39 bands (See Supplementary Material 2: Band division), parting from  
296 the original 21 wavelength bands from the atmospheric radiation model (Berger and  
297 Bathiebo, 1989).

298 An emissivity value of zero or non-zero was assigned to each band; fifteen wavelength  
299 combinations were proposed. To establish the bandwidth for an ideal material, the non-  
300 zero emissivity values of the theoretical materials were selected to be centered in the  
301 transparency window of the atmosphere (infrared emission), shown in Figure 5.

302 Similarly, for the visible region, the emissivity values were centered in the region with  
303 the highest solar irradiance. The band combinations resulted in the 15 theoretical  
304 materials (M1-M15) shown in Figure 4. Moreover, to quantify the impact of the  
305 emissivity value, six emissivity values (1, 0.9, 0.8, 0.7, 0.5, and 0.25) were assigned to  
306 the non-zero value, resulting in a total of 90 theoretical materials.

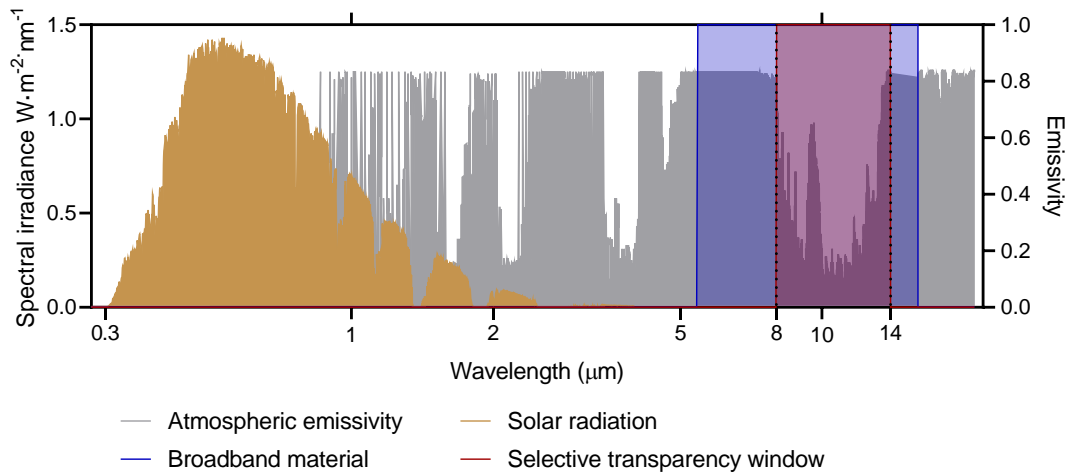




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308 *Figure 4. Emissivity of the theoretical materials (M1-M15) resulting from combining emissivity*

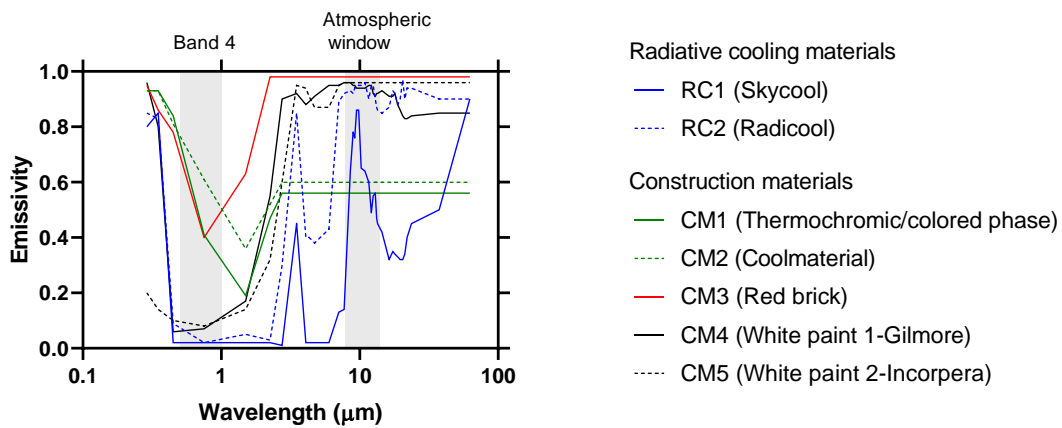
309 *values of 1 and 0 in the 39 wavelength bands.*



310

311 *Figure 5. Solar spectrum, atmospheric emissivity, and ideal material spectra.*

312 The performance of the theoretical radiative cooling materials (M1-M15) was compared  
 313 with existing radiative cooling materials (Skycool (RC1) and Radicool (RC2)) and other  
 314 construction materials (CM1-CM5 e.g. white paints, brick, and coolmaterials) for the  
 315 same boundary conditions. This provides a better understanding of the potential  
 316 benefits and ways to improve these materials.

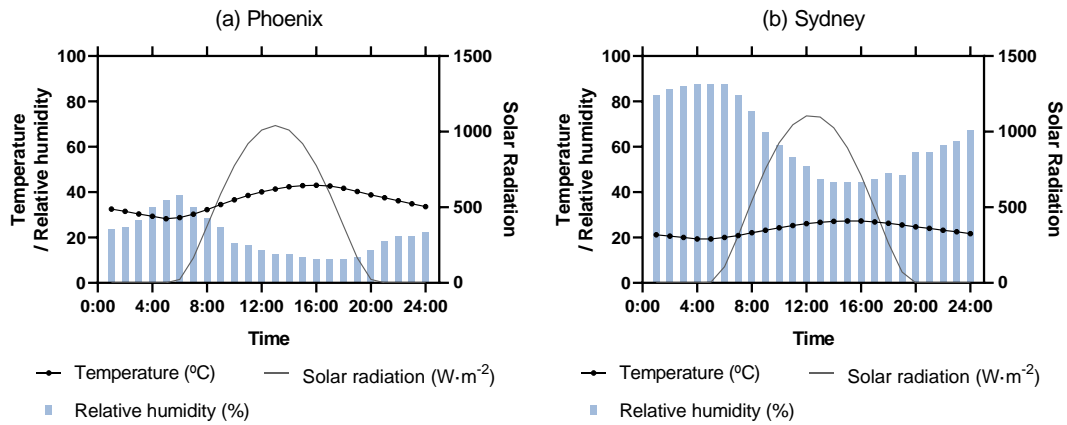


317

318 *Figure 6. Emissivity comparison of existing radiative cooling materials (RC) and construction*  
 319 *materials (CM).*

320 The sample's thermal response was calculated in two different cities, Phoenix (hot and  
 321 dry) and Sydney (mild and humid) during the summer solstice, on 21<sup>st</sup> June and 21<sup>st</sup>

322 December, respectively. The climates of the selected cities obtained from Meteonorm  
 323 (*Meteonorm 7*, 2017) represent completely clear skies and are shown in Figure 7.



324

325 *Figure 7. Hourly climatic parameters in summer solstice for Phoenix and Sydney.*

326 **3. Results**

327 Two different simulation conditions were considered: a highly insulated and a very  
 328 conductive surface (Table 2).

329 *Table 2: Material substrate for two conditions*

Background conditions	Material	Thickness	Thermal conductivity	Density	Specific Heat
1. Insulated	Insulation	0.15 m	0.0001 W/m·K	0 kg/m <sup>3</sup>	1000 J/kg·K
2. Conductive	Metallic sheet	0.005 m	400 W/m·K	0 kg/m <sup>3</sup>	1000 J/kg·K

330

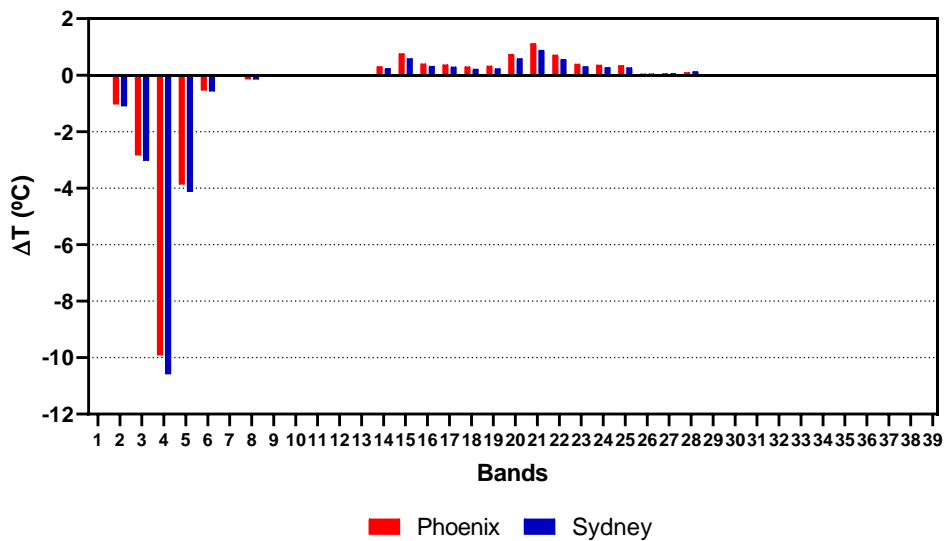
331 **3.1. Performance of the samples over a highly insulated surface**

332 The first thermal scenario considers the material insulated entirely on the bottom side  
 333 to have almost no conductivity; there is negligible heat transfer by conduction.

334 Therefore, this condition can be regarded as almost adiabatic. Below the insulation, the  
 335 temperature was 25 °C, and the exterior convective heat transfer coefficient was 20

336  $W \cdot m^{-2} \cdot K^{-1}$ . In this case, the resulting variable of interest is the surface temperature  
 337 reached.

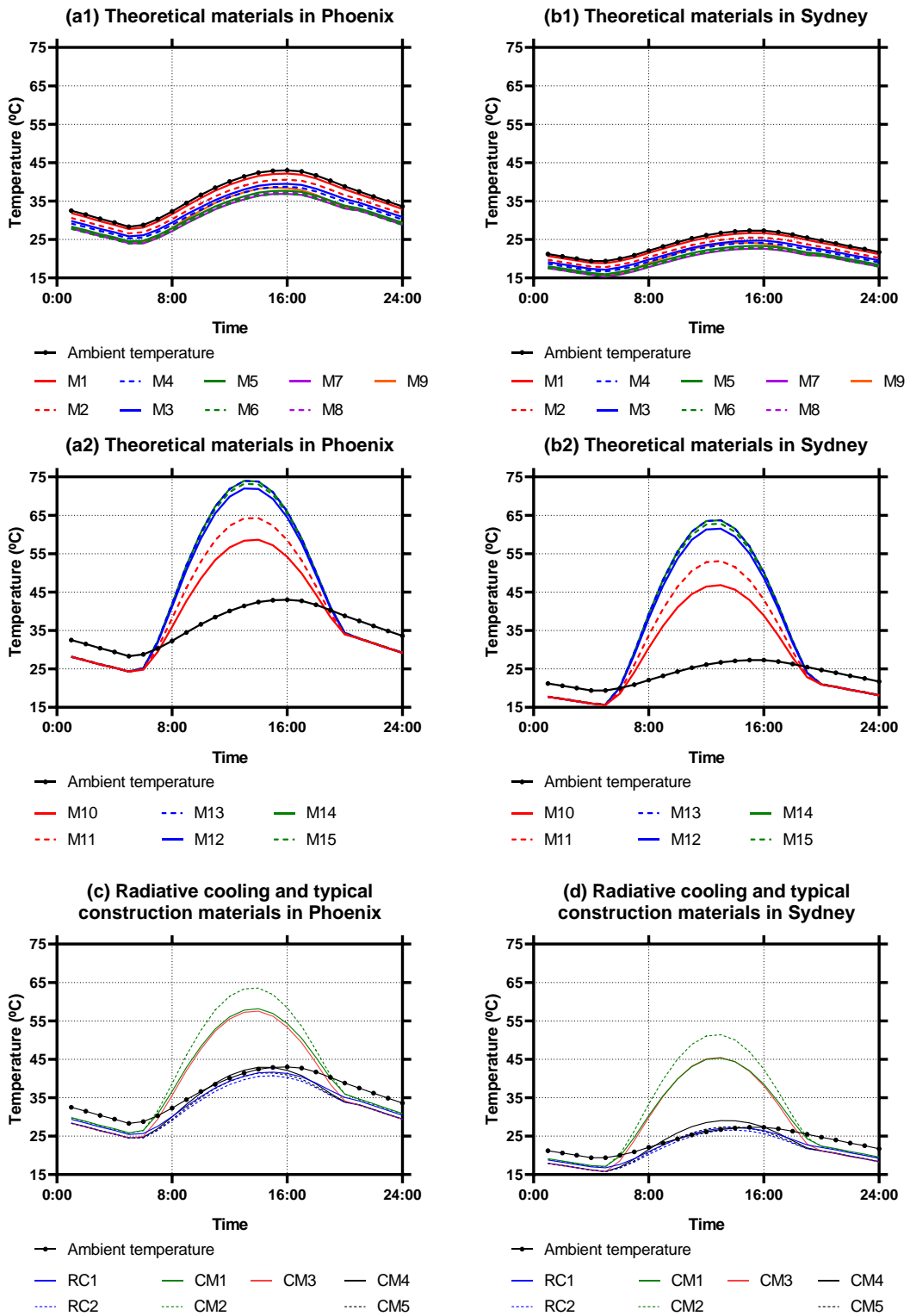
338 To study the effect of each of the 39 bands, the emissivity value in the selected band  
 339 was 1 and 0 in the rest of the 38 bands. As Figure 8 shows, from “Band 2” (0.3-0.4  $\mu m$ )  
 340 to “Band 7” (2.5-3  $\mu m$ ), having an emissivity of 1 leads to heat gains, especially in “Band  
 341 4” (0.5-1  $\mu m$ ), where the material is 10.57  $^{\circ}C$  and 9.90  $^{\circ}C$  hotter than the ambient  
 342 temperature in Sydney and Phoenix, respectively. On the other hand, “Band 15” (8.29-  
 343 8.82  $\mu m$ ) and “Band 20” (9.98-10.50  $\mu m$ ) to “Band 22” (11.33-11.95  $\mu m$ ) have a high  
 344 impact on the heat losses, reaching a reduction of 1.13  $^{\circ}C$  in Phoenix and 0.88  $^{\circ}C$  in  
 345 Sydney when the emissivity of “Band 21” (10.5-11.325  $\mu m$ ) equals 1. Absorbing heat in  
 346 the solar wavelengths has a more significant effect than the emissive power inside the  
 347 atmospheric window, as can be seen in Figure 8.



348  
 349 *Figure 8. Contribution of each band’s emissivity to the average temperature difference. Positive*  
 350 *values are bands that achieved sub-ambient cooling and negative values are those that*  
 351 *reached higher than ambient temperatures.*

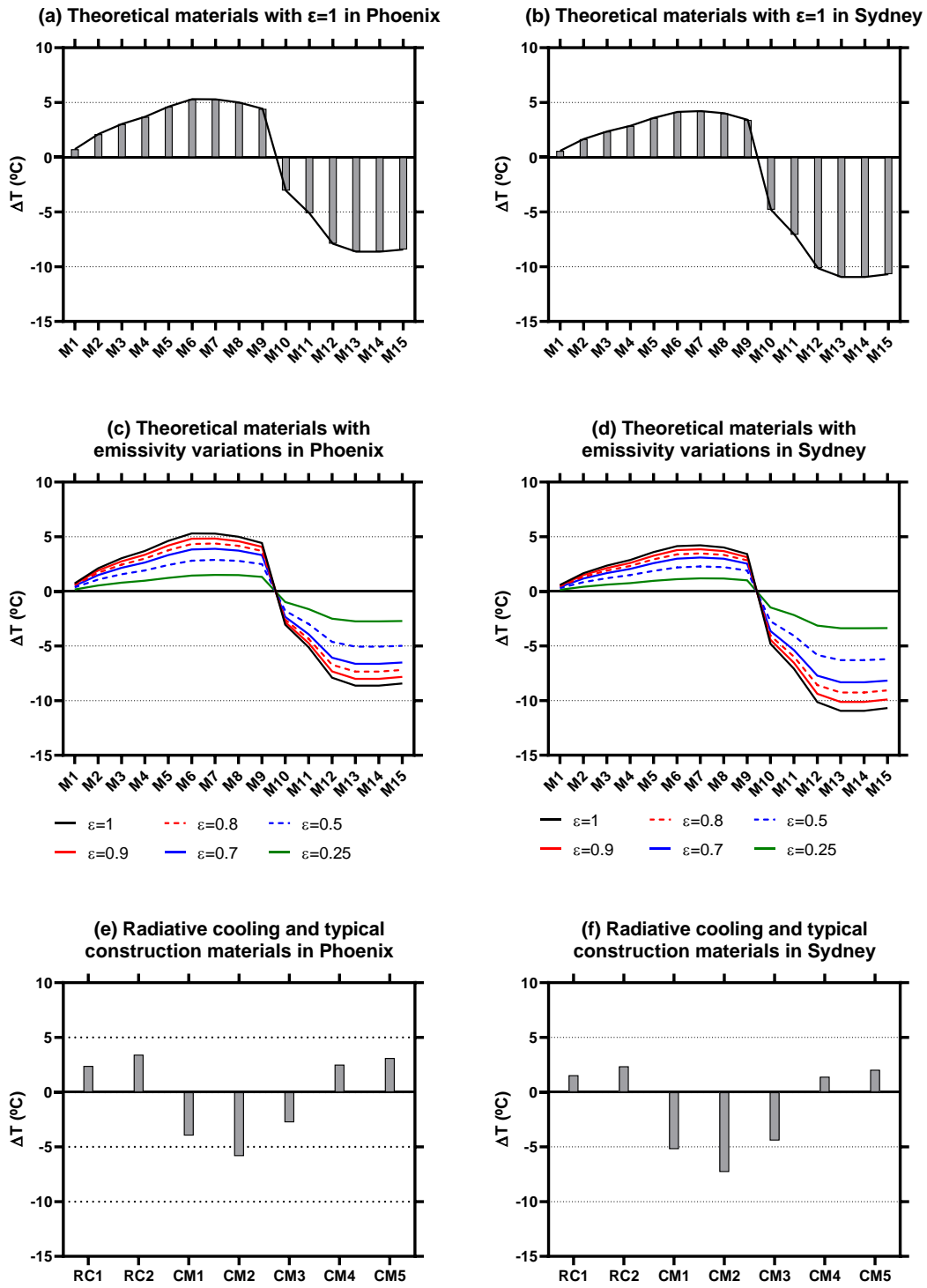
352 Once the effect of each band was known, the surface temperature of the 15 theoretical  
353 materials was calculated with the six possible emissivity values and the existing  
354 materials (Figure 9). The results for the theoretical materials were divided into two  
355 groups: those that achieved sub-ambient cooling during the day (M1-M9, Figure 9 a1,  
356 b1), and the ones that reached higher than ambient temperatures (M10-M15, Figure 9  
357 a2, b2). All the materials achieved higher temperature reductions in Phoenix than in  
358 Sydney. M6 achieved a 5.29 °C reduction in Phoenix and M7 a reduction of 4.20 °C in  
359 Sydney (see Figure 10 a1, b1), whereas RC2 reached a mean temperature drop of  
360 3.42 °C in Phoenix and 2.36 °C in Sydney. CM5 achieved a very similar temperature  
361 reduction, 3.12 °C, and 2.05 °C, respectively. The thermochromic paint (CM1), the cool  
362 material (CM2), and the red brick (CM3) did not achieve sub-ambient temperatures  
363 during the day (Figure 9 c, d).

364 Lowering the emissivity of the theoretical materials led to a reduction in the attained  
365 surface temperature, as seen in Figure 10 (c, d); however, a material with an emissivity  
366 of 0.25 in the atmospheric window and 0 outside achieved a temperature reduction of  
367 1.51 °C in Phoenix and 1.17 °C in Sydney. During the night, all the studied materials,  
368 theoretical and existing, achieved sub-ambient cooling.



369

370 *Figure 9. Hourly surface temperature achieved by the materials in Phoenix and Sydney.*



371

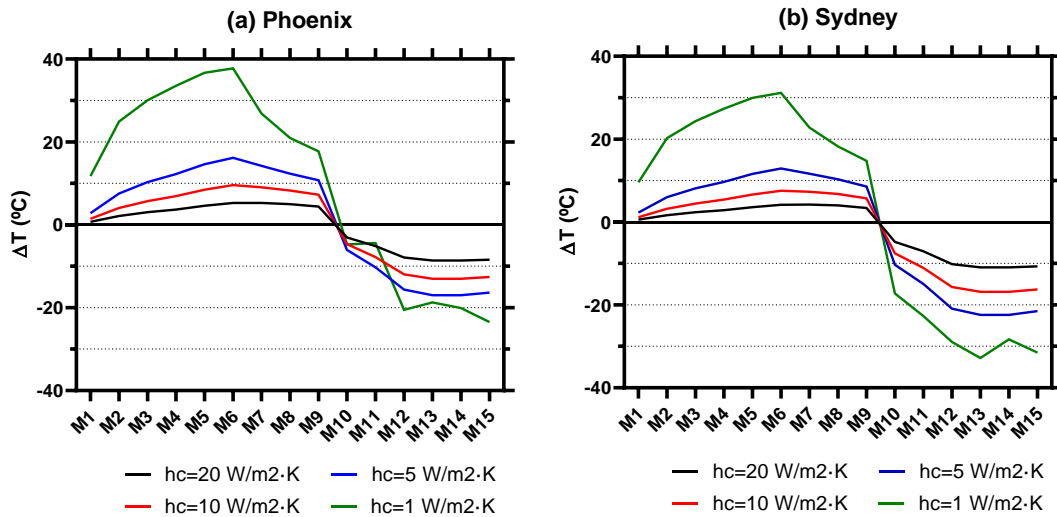
372 Figure 10. Difference between mean ambient and surface temperature for theoretical materials  
 373 (M1-M15), radiative cooling materials (RC1-RC2) and typical construction materials (CM1-CM5)

374 *in Phoenix and Sydney. Positive values are materials that achieved sub-ambient cooling and*  
375 *negative values higher than ambient temperatures.*

376 Figures 9 and 10 above show summarized behavior and a comparison of the simulated  
377 materials considering a highly insulated condition. If the objective is to achieve the  
378 minimum surface temperature, the ideal material is M6 for Phoenix and M8 for Sydney.  
379 Nevertheless, in both cases (mainly for Phoenix), the difference between M6 and M8 is  
380 low. Therefore, the ideal material should have an emissivity of 1 approximately in the  
381 band between 5 and 17 $\mu$ m. The emissivity in the visible region has a powerful impact  
382 on the behavior of the material since the infrared emission cannot be compensated by  
383 solar absorption. The emissivity of the white paints (CM4 and CM5) was very similar to  
384 “Skycool” and “Radicool” (RC1 and RC2), and therefore their thermal behavior is  
385 similar to radiative cooling materials.

386 Finally, in order to study the effect of the convective coefficient, the mean surface  
387 temperature achieved by M1 to M15 was calculated and is shown in Figure 11. When  
388 the convection is reduced, Phoenix (M6, 37 °C) had the potential to achieve a lower  
389 sub-ambient temperature than Sydney (M6, 31 °C). On the other hand, a lower  
390 convective rate led to a higher surface temperature in Sydney (M15, 31 °C) than in  
391 Phoenix (M15, 23 °C).





392

393 *Figure 11. Difference between mean ambient and surface temperature for theoretical materials*  
 394 *(M1-M15) with different convective values in Phoenix and Sydney. Positive values mean sub-*  
 395 *ambient cooling and negative values higher than ambient temperatures.*

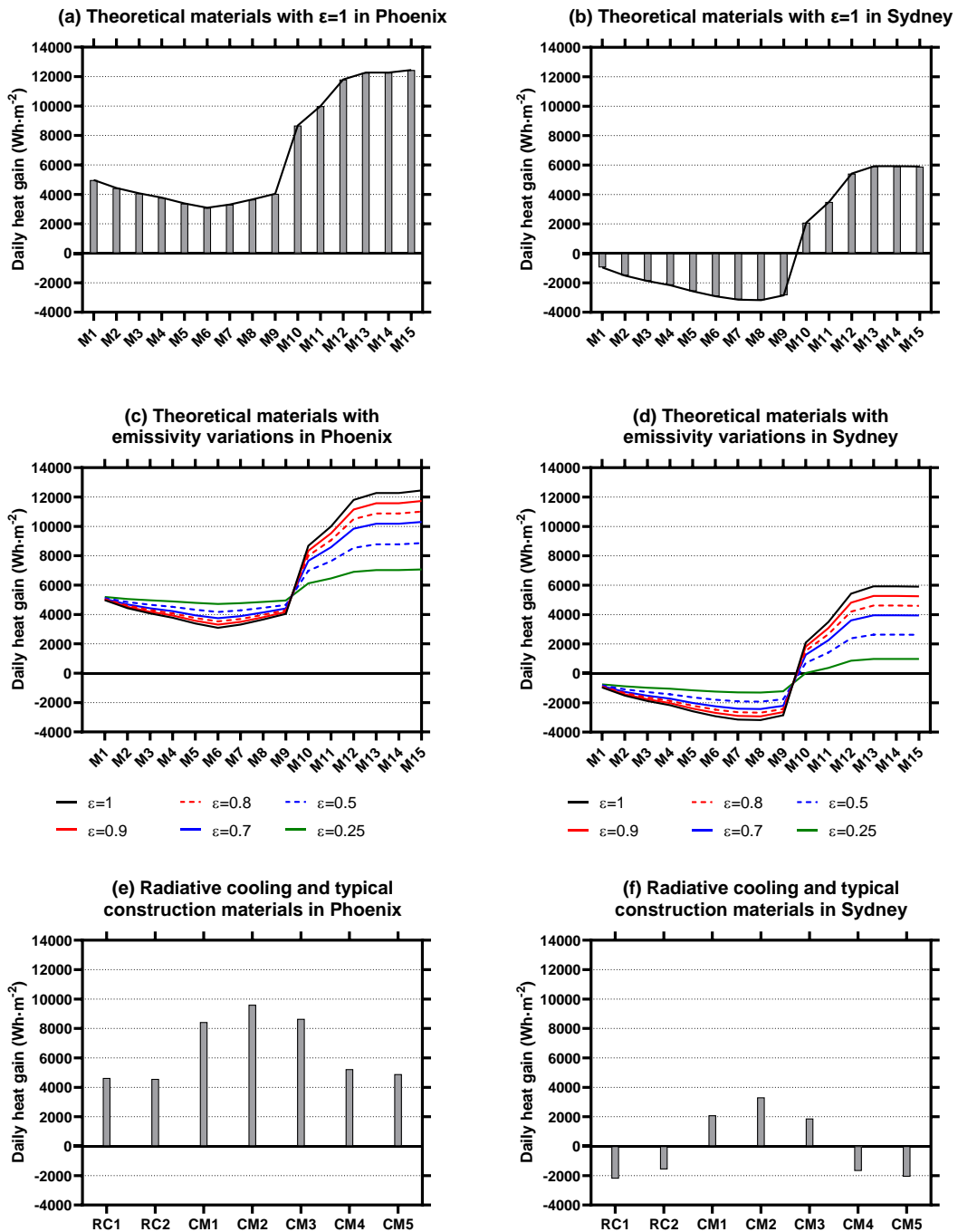
### 396 **3.2. Performance of the samples over a conductive surface**

397 In the second scenario, materials are placed on top of a very conductive surface with  
 398 no insulation. This scenario mimics the idea of having a fluid or a heat source at a  
 399 constant temperature under the surface and calculates the cooling potential. Below the  
 400 material, the temperature is 25 °C, the exterior heat transfer coefficient is 25 W·m<sup>-2</sup>·K<sup>-1</sup>,  
 401 and the interior heat transfer coefficient is 1000 W·m<sup>-2</sup>·K<sup>-1</sup>.

402 The daily heat gains were calculated for all the materials in both cities and are  
 403 represented in Figure 12. In the case of Phoenix (Figure 12 a), using a very conductive  
 404 material leads to heat gains since the mean ambient temperature that day is 36.19 °C.  
 405 Nevertheless, the theoretical material M6 with an emissivity of 1 achieves the lowest  
 406 heat gain of 3091 Wh·m<sup>-2</sup>, followed by M7 (3306 Wh·m<sup>-2</sup>) and M5 (3384 Wh·m<sup>-2</sup>).  
 407 Among the existing materials, the behavior of RC2 (4578 Wh·m<sup>-2</sup>) and CM4 (5240  
 408 Wh·m<sup>-2</sup>) is closer to that of RC1 (4645 Wh·m<sup>-2</sup>) and CM5 (4897 Wh·m<sup>-2</sup>). The mean  
 409 ambient temperature in Sydney for that day is 23.6 °C (Figure 12 b). Materials M1-M9

410 achieved a substantial heat loss: theoretical material M8 attained the highest heat loss  
411 of  $-3176 \text{ Wh}\cdot\text{m}^{-2}$  followed by M7 ( $-3140 \text{ Wh}\cdot\text{m}^{-2}$ ), and M6 ( $-2916 \text{ Wh}\cdot\text{m}^{-2}$ ), when the  
412 emissivity value is 1 (Figure 12 b). Among the existing materials, the highest heat  
413 losses correspond to RC1 ( $-2212 \text{ Wh}\cdot\text{m}^{-2}$ ) and CM5 ( $-2077 \text{ Wh}\cdot\text{m}^{-2}$ ). Contrary to the  
414 situation in Phoenix, despite having similar optical properties to RC1 and CM5,  
415 materials RC2 and CM4 achieved values that were around  $700 \text{ Wh}\cdot\text{m}^{-2}$  lower.

416 In the case of Phoenix (Figure 12 c), reducing the emissivity of the theoretical materials  
417 M1 to M9 leads to higher heat gains. Material M6 attains the best behavior since it has  
418 the lowest heat gains. Once the materials start to absorb in the solar wavelengths, the  
419 higher the emissivity, the greater the heat gains are. In the case of Sydney (Figure 12  
420 d), reducing the emissivity of the theoretical materials leads to lower heat losses for M1  
421 to M9. Material M8 attains the best behavior as it has the highest heat losses.

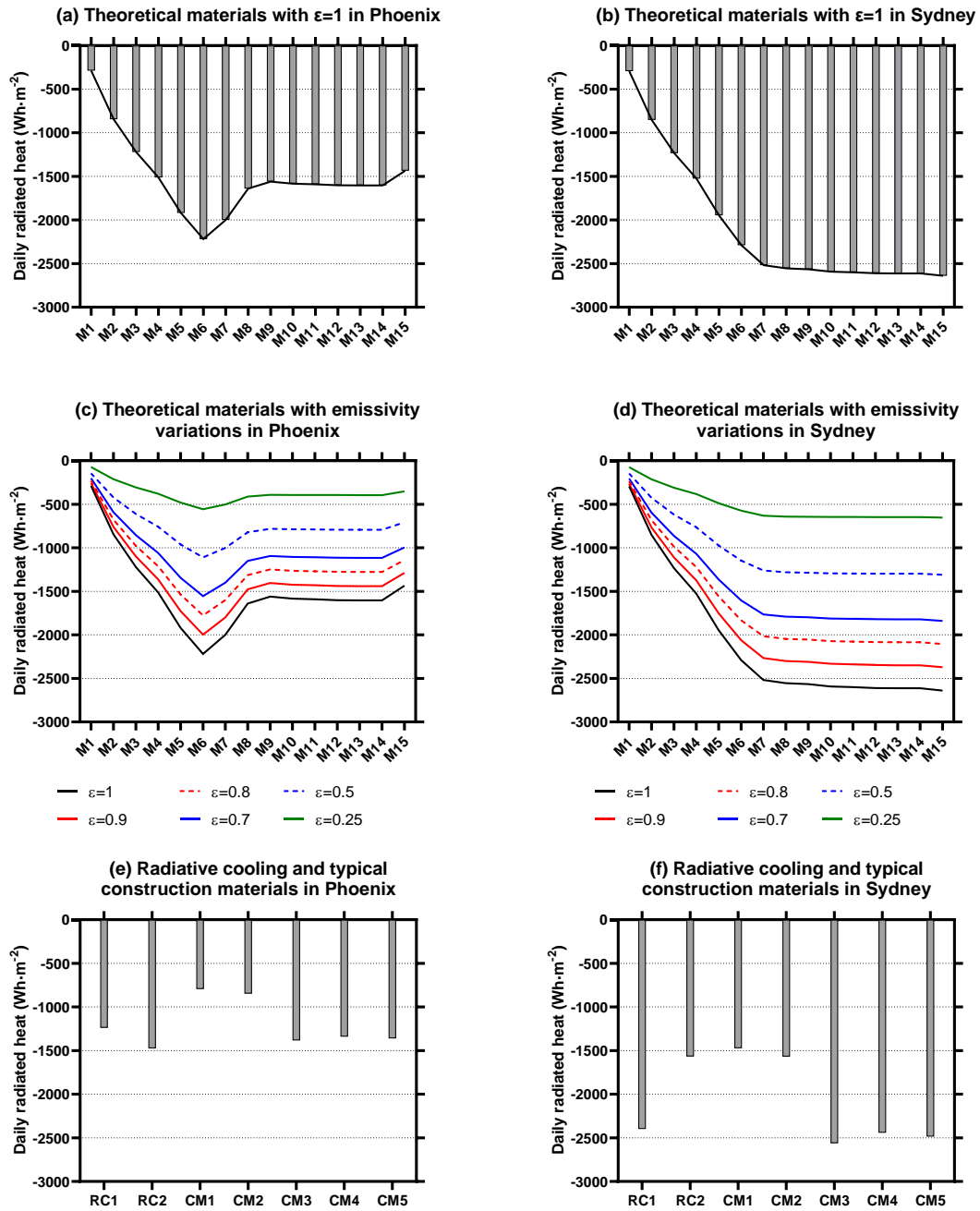


422

423 Figure 12. Daily gains or losses for theoretical materials (M1-M15), radiative cooling materials  
 424 (RC1-RC2) and typical construction materials (CM1-CM5) in Phoenix and Sydney. Positive  
 425 values are heat gains and negative are heat losses.

426 The daily accumulated radiated heat of each surface is represented in Figure 13. As  
 427 can be seen, the potential is higher in Sydney than in Phoenix due to the difference in

428 the ambient temperatures of both cities; that the bottom surface is at 25 °C hinders  
429 enormously the cooling ability in Phoenix, where M6 attains the highest radiation  
430 power,  $-2218 \text{ Wh}\cdot\text{m}^{-2}$ . (Figure 13 a). The difference between using one of the  
431 theoretical materials and the already developed ones is substantial, the radiated heat  
432 almost halving in the latter (Figure 13 e). RC2 achieved  $-1475 \text{ Wh}\cdot\text{m}^{-2}$  and CM5  $-1359$   
433  $\text{Wh}\cdot\text{m}^{-2}$ . In the case of Sydney (Figure 13 b), from M7 ( $-2517 \text{ Wh}\cdot\text{m}^{-2}$ ) onwards, all the  
434 materials achieve a similar radiation power. In this case, RC2, and RC5 achieve  $-2398$   
435  $\text{Wh}\cdot\text{m}^{-2}$ ,  $-2484 \text{ Wh}\cdot\text{m}^{-2}$ , respectively. In both cities, lowering the emissivity leads to  
436 lower radiated heat (Figure 13 c, d). When the exchange temperature is higher, the  
437 theoretical radiative cooling materials perform better, as shown in Phoenix.

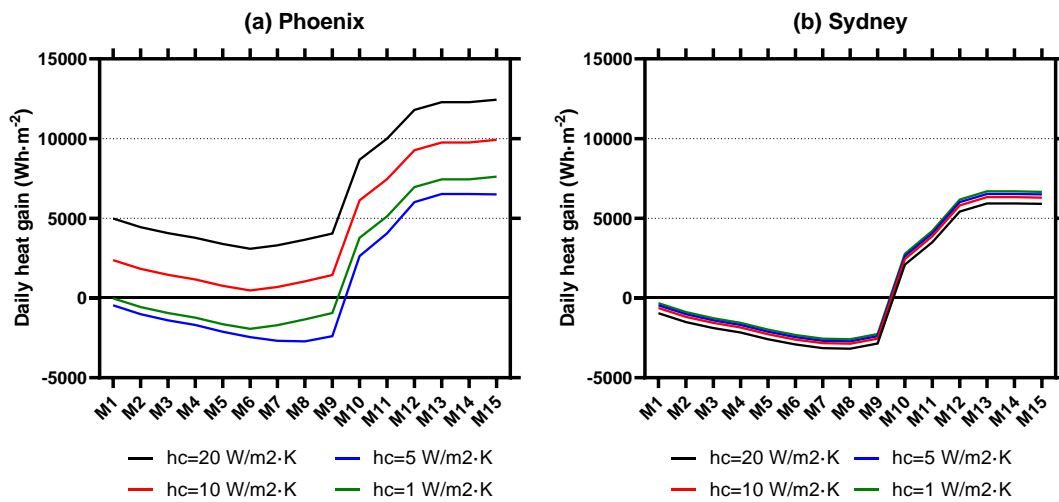


438

439 *Figure 13. Daily radiated heat for theoretical materials (M1-M15), radiative cooling materials*  
 440 *(RC1-RC2) and typical construction materials (CM1-CM5) in Phoenix and Sydney. Positive*  
 441 *values are heat gains and negative are heat losses.*

442 Finally, to study the effect of the convective coefficient, the thermal gains for materials  
 443 M1 to M15 were calculated and are shown in Figure 14. As mentioned above, the  
 444 mean ambient temperature of Phoenix is higher than the interior temperature

445 considered. Therefore, in this case, the higher the convection, the lower the cooling  
 446 capacity in Phoenix; the air temperature heats the surface leading to considerable heat  
 447 gains. Convection plays a less significant role in Sydney since the interior temperature  
 448 is closer to the ambient temperature.



449

450 *Figure 14. Daily gains or losses for theoretical materials (M1-M15) with different convective*  
 451 *values in Phoenix and Sydney. Positive values are heat gains and negative are heat losses.*

#### 452 4. Conclusions

453 This paper analyzes the sensitivity of the performance of daytime radiative cooling  
 454 materials to different spectral selectivity configurations, type of application, and  
 455 location. The results presented in this paper suggest that the kind of application (active  
 456 or passive) is a determinant factor in the design of radiative cooling materials. A  
 457 material that performs well in a dry climate as a passive solution could perform poorly  
 458 as an active solution. When used as an active solution, the operating temperature and  
 459 climate should be carefully studied.

460 The radiation spectrum was divided into 39 bands and the contribution of each band  
 461 was calculated. The most critical bands regarding heat absorption are band 4 (0.5-1  
 462  $\mu\text{m}$ ) followed by band 5 (1-2  $\mu\text{m}$ ) and band 3 (0.4-0.5  $\mu\text{m}$ ). A material that solely emits

463 in band 4 reaches a surface temperature up to 10.6 °C higher than the ambient  
464 temperature in Sydney and 9.9 °C in Phoenix. Therefore, it is important to achieve high  
465 reflectivity in the 0.5-1 μm region. The emissivity values should be especially high in  
466 Bands 20-22 (9.98-11.95 μm). Combining the 39 bands, a total of 15 theoretical  
467 materials with 6 different emissivity values were proposed and compared to existing  
468 daytime radiative cooling materials and typical construction materials. As many authors  
469 have previously mentioned, the results of the daytime radiative cooling materials could  
470 not be directly compared. However, the present research has made it possible to  
471 compare under the same conditions the results of theoretical materials (M1-M15) with  
472 two of the most innovative radiative cooling materials in recent years, “Skycool” RC1  
473 and “Radicool” RC2.

474 Modifying the materials’ optical properties leads to a substantial change in the heat  
475 gains or losses in an active system and the surface temperature reached as a passive  
476 application. The most suitable optical spectrum for a material is determined by the  
477 climate of each location (Sydney and Phoenix in this study) and the application type  
478 (boundary conditions). The highly insulated condition was more beneficial in Phoenix,  
479 where the theoretical materials achieved (M6, 5.30 °C) a higher sub-ambient cooling  
480 temperature than in Sydney (M6, 4.21 °C). On the other hand, the materials that did not  
481 present sub-ambient cooling during the day (M10-M15) showed worse behavior in  
482 Sydney than in Phoenix due to the higher humidity. M14 reached a surface  
483 temperature 10.93 °C higher than the ambient temperature in Sydney and 8.63 °C  
484 higher than in Phoenix. Using a radiative cooling material over a very conductive  
485 surface requires a different approach. In Sydney, a broader spectrum outside the  
486 atmospheric window was more beneficial than one solely within the transparency  
487 window. The theoretical material M8 achieved the highest daily heat losses (-3176  
488 Wh·m<sup>-2</sup>). In this case, the existing materials RC1 and CM5 are good alternatives to the  
489 theoretical materials. In Phoenix, on the other hand, restricting the emissivity to the

490 atmospheric transparency window resulted in better behavior. Theoretical material M6,  
 491 with an emissivity of 1, achieved the lowest heat gain of 3091 Wh·m<sup>-2</sup>.

492 If the average temperature increase in urban areas reaches the predicted 4 to 5 °C,  
 493 daytime radiative cooling materials are great candidates to counteract it. Radiative  
 494 cooling is of special interest in cities suffering from the UHI effect since the heat  
 495 accumulated during the day will be evacuated to outer space instead of to the streets,  
 496 alleviating the heat buildup in cities and breaking the vicious cycle of increasing cooling  
 497 demand. However, more research is necessary to determine how to apply this to the  
 498 built environment. The impact of building radiation on the ability of these materials to  
 499 cool down should be studied in more depth.

500 *Table 3: Summary of simulations results.*

Phoenix		Sydney	
Insulated			
Material	$\Delta T$ (°C) difference	Material	$\Delta T$ (°C) difference
M6	5,30 °C	M7	4,21 °C
RC2 Radicool	3,42 °C	RC2 Radicool	2,36 °C
RC1 Skycool	2,40 °C	RC1 Skycool	1,55 °C
CM5 White paint 2	3,12 °C	CM5 White paint 2	2,04 °C
CM4 White paint 1	2,52 °C	CM4 White paint 1	1,41 °C
Conductive			
Material	Daily heat gains	Material	Daily heat gains
M6	3091 W·m <sup>-2</sup>	M8	-3176 W·m <sup>-2</sup>
RC2 Radicool	4578 W·m <sup>-2</sup>	RC2 Radicool	-2213 W·m <sup>-2</sup>
RC1 Skycool	4644 W·m <sup>-2</sup>	RC1 Skycool	-1574 W·m <sup>-2</sup>
CM5 White paint 2	4897 W·m <sup>-2</sup>	CM5 White paint 2	-2077 W·m <sup>-2</sup>
CM4 White paint 1	5240 W·m <sup>-2</sup>	CM4 White paint 1	-1689 W·m <sup>-2</sup>

501

## 502 Declaration of interest

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