Supplement of 

Development of a continuous UAV-mounted air sampler and application to the quantification of CO₂ and CH₄ emissions from a major coking plant 

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Section SI-1. Evaluation of CO₂ emissions through carbon material balance

Figure S1 illustrates the coking process flow and the structure of a coking oven. Coking is a process in which coking coal is heated in an oxygen-free environment to produce coke, a high-carbon metallurgical coke used in steel production. The process takes place in a coke oven, where coking coal is heated at temperatures exceeding 1000 °C, driving off all volatile components of the coal and leaving behind coke. Coke oven gas (COG), the byproduct of coking, is mainly composed of the components listed in Table S1 (Razzaq et al., 2013). This gas is recovered and mostly reused as fuel in firing the coke oven to maintain the high temperatures needed for coking (Zhang, 2019), reducing/eliminating the need for external fuel sources. However, burning COG generates CO₂, which is the primary waste gas emitted.

![Diagram of coking process](image)

**Figure S1.** The conceptual coking process flow and the structure of the coking oven

**Table S1.** Main carbon constitutes and their corresponding compositions (vol%) in coke oven gas

<table>
<thead>
<tr>
<th>Component</th>
<th>Content</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₄</td>
<td>23~27%</td>
</tr>
<tr>
<td>CO₂</td>
<td>1.5~3%</td>
</tr>
<tr>
<td>CO</td>
<td>5~8%</td>
</tr>
<tr>
<td>C₂H₄</td>
<td>2~4%</td>
</tr>
</tbody>
</table>
The main products of coking are coke, COG, and slag. In Chinese coking plants (Zhang, 2019), typically half of the produced COG is used as fuel in firing the coke oven, while a small portion is recycled for producing chemical products, and the rest is either leaked (5 %) (Hein, 2012) or an unknown fraction is directly released into the atmosphere. The Shagang coking plant has implemented a process for recycling slag. This process involves reusing the slag, which is generated during the coking process, as part of the fuel for the coking oven. As a result, the carbon in the slag is similarly oxidized into CO$_2$ and subsequently released into the atmosphere as well. Therefore, the CO$_2$ emissions ($E_{CO_2}$) mainly come from the combustion of COG ($E_{combustion-COG}$), the combustion of slag ($E_{combustion-slag}$), and the direct release of COG ($E_{release}$):

$$E_{CO_2} = E_{combustion-COG} + E_{combustion-slag} + E_{release}$$  \hspace{1cm} (1)$$

Based on the carbon material balance, the amount of carbon in COG combusted in the coking oven ($C_{combustion-COG}$) during the coking process can be derived from material balance:

$$C_{combustion-COG} = \alpha(C_{coal} - C_{coke} - C_{slag})$$  \hspace{1cm} (2)$$

where $C_{coal}$ is the amount of carbon in coal, $C_{coke}$ the amount in coke, and $C_{slag}$ the amount in slag. $\alpha$ is the fraction of COG used in firing the coking oven and is 0.5 based on operation data at the Shagang coking plant. The measured coking plant consists of two coke oven batteries, each with its own stack. According to the faculty at the coking plant, each coke oven battery produced 127.8 t coke hr$^{-1}$, thus totalling 255.6 t coke hr$^{-1}$ ($p_{coke}$) between the two batteries during the UAV measurement period with a coke yield of 78.5 %. The coking batteries are in continuous operation and the coking time (the time from charging the coal to pushing of coke out of the oven) is 26 hours. Thus a 30-minute flight is a suitable length of time for measurement and material balance analysis. The carbon content of coking coal typically ranges from 80 % to 87 % (Dai et al., 2022). Assuming an average value of 83.5 %, the total amount of carbon in the coking coal used by the coking plant per hour can be calculated as:

$$C_{coal} = \frac{p_{coke}}{78.5\%} \times 83.5\% = 272 \ t \ C \ hr^{-1}$$  \hspace{1cm} (3)$$

According to the US EPA, metallurgical coke has a carbon content of 82 % to 87 % (U.S. Environmental Protection Agency, 2008). If an average value of 84.5 % is taken, the produced carbon in the coke during the coking process can be calculated as:

$$C_{coke} = p_{coke} \times 84.5\% = 216 \ t \ C \ hr^{-1}$$  \hspace{1cm} (4)$$

Generally speaking, the yield of slag is 0.05 % to 0.07 % of the coal charged for coking process (an
average value of 0.06 % is taken here), and that the carbon content of slag is around 80 % (Li, 2022). Thus, the total amount of carbon in the produced slag can be calculated as:

\[ C_{\text{slag}} = \frac{p_{\text{coke}}}{78.5\%} \times 0.06\% \times 80\% = 0.16 \text{ t C hr}^{-1} \]  

(5)

by substituting Eq. (3), (4) and (5) into Eq. (2), the mass of carbon in the combusted COG \((C_{\text{combustion-COG}})\) during the coking process is calculated to be 27.9 t hr\(^{-1}\). Thus, \(E_{\text{combustion-COG}}\) and \(E_{\text{combustion-slag}}\) can be calculated respectively:

\[ E_{\text{combustion-COG}} = C_{\text{combustion-COG}} \times \frac{M_{\text{CO}_2}}{M_{C}} = 102.3 \text{ t CO}_2 \text{ hr}^{-1} \]  

(6)

\[ E_{\text{combustion-slag}} = C_{\text{slag}} \times \frac{M_{\text{CO}_2}}{M_{C}} = 0.59 \text{ t CO}_2 \text{ hr}^{-1} \]  

(7)

where \(M_{\text{CO}_2}\) and \(M_{C}\) is the molar mass of \(\text{CO}_2\) and the atomic mass of carbon, respectively.

As certain amount of \(\text{CO}_2\) is directly released into the atmosphere along with COG, the carbon mass in the released and measured \(\text{CO}_2\) \((C_{\text{release}})\) can be derived as:

\[ C_{\text{release}} = \beta(C_{\text{coal}} - C_{\text{coke}} - C_{\text{slag}}) \times \frac{\varphi_{\text{CO}_2}}{\varphi_{\text{CO}_2} + \varphi_{\text{CH}_4} + \varphi_{\text{CO}} + 2\varphi_{\text{C}_2\text{H}_4}} = 0.16 \text{ t hr}^{-1} \]  

(8)

where \(\beta\) is fraction of COG that is directly released into the atmosphere, taken to be 0.05 as described above, \(\varphi_{\text{CO}_2}\), \(\varphi_{\text{CH}_4}\), \(\varphi_{\text{CO}}\), and \(\varphi_{\text{C}_2\text{H}_4}\) are the volume fractions for the main constituents in COG (Table S2). Thus, the corresponding \(\text{CO}_2\) emissions from the direct release of COG \((E_{\text{release}})\) can be derived as:

\[ E_{\text{release}} = C_{\text{release}} \times \frac{M_{\text{CO}_2}}{M_{C}} = 0.59 \text{ t CO}_2 \text{ hr}^{-1} \]  

(9)

by substituting Eq. (6), (7) and (9) into Eq. (1), the total \(\text{CO}_2\) emissions \((E_{\text{CO}_2})\) from the full coking process is calculated to be 103 t \(\text{CO}_2\) hr\(^{-1}\).

Taking into account the variation in carbon content found in both coking coal and coke, the variation in the fraction of COG used as fuel (assuming a range of 0.4 to 0.6), the uncertainty of slag yield, as well as the range in the volume fraction of the main components of COG, the total uncertainty range of \(\text{CO}_2\) released into the atmosphere during the coking process can be estimated to be 31 % by the equation below:

\[ \delta^2 = \delta^2_{\text{coal}} + \delta^2_{\text{coke}} + \delta^2_{\text{as fuel}} + \delta^2_{\text{slag yield}} + \delta^2_{\text{volume fraction}} \]  

(10)
Thus, the total amount of CO$_2$ released into the atmosphere during the full coking process estimated from the coke production data is 103 ± 32 t CO$_2$ hr$^{-1}$, which is consistent with the CO$_2$ emission results (110 ± 18 t CO$_2$ hr$^{-1}$, see main text) from the current UAV-measurements, as shown in Figure S2.

**Figure S2.** Comparison between the measured and estimated CO$_2$ emission rates from material balance

**References**


