

Overall – the method appears potentially impressive from the data presented here, but the quality of the paper is let down by some imprecise writing and concerns around cherry picking of data for presentation. It feels that it needs some sort of controlled release validation to be fully compelling as a go-to method of quantification. Although probably out of scope of this work (given that this is part of the CoMET special issue) this quantification method should be tested with data collected at blind control release experiments to ensure that it is truly capable of describing real emissions, and therefore able to correctly understand the “real” uncertainty rather than the mathematical uncertainty within these quantifications. As it stands, I feel the authors need to explain some parts in more detail to alleviate concerns and make some significant improvements to the quality of writing throughout.

It will be suitable for full publication once the problems are ironed out.

Dear reviewer, thank you for all your kindly comments, which are very helpful in improving the quality of our manuscript. We think it is very reasonable to use the measured quantitative data to verify our proposed method, which can help readers to have more confidence in the presented calculation results. However, it is difficult to conduct control experiments for ventilation shafts in methane mines. Therefore, we use similar controllable experiments to verify our method. In our latest modified manuscript, we evaluate the performance of GA-IPPF in three release-controlled experiments: the vehicle-mounted CH<sub>4</sub> dataset of EPA, the CH<sub>4</sub> release dataset with Aircore system, and the SO<sub>2</sub> dataset measured by ground-based in-situ network.

The results of the above experiments showed the high accuracy of GA-IPPF, and the reconstructed methane diffusion results are consistent with actual measured ones. These control experiments can not only be used as a verification part, but also as a proof of whether GA-IPPF is applicable to the quantification of emission sources by other measurement systems.

As you mentioned, since this is the COMET special issue, the main purpose of our paper is to highlight the performance of GA-IPPF in assessing methane emissions in COMET. Thus, the evaluation of release-controlled experiments are presented in Discussion section.

Finally, we are very grateful for your approval of our manuscript, and we have organized and answered your comments one by one. Words in blue color are your comments, and our responses are in black color words. Detailed responses are as follows:

### **Major thoughts**

1. The volatility of emissions from mining operations is not surprising given the variable conditions and operational mine venting. The aircore system is a well established method of data collection and it is good to see it being able to capture the emissions nice and clearly. I have concerns about the data quality though for being able to fully resolve the mass balance with any certainty. In Figure 3a, the plume is only intercepted once along a single transect and is not fully bounded with background air to the South. In Fig 3b, there appears to be two hotspots, with the main lofted plume not resolved to the West at all. This is one of the downsides of not measuring inflight and only being able to measure post-flight. Whilst the uncertainties of the GA-IPPF are low, it needs

to be stated that we do not know the error in the accuracy of the method as we have no pre-defined truth to compare to.

Thank you for this suggestion. First, when we used UAV-based Aircore system to collect the diffusion of CH<sub>4</sub> leakage, at a certain height, the UAV will fly laterally to ensure that Aircore system would detect larger value in the track. Because the high concentration can represent the diffusion characteristics of the emission source. GA-IPPF would ensure that a global optimal solution of unknow parameters can be obtained in Gaussian dispersion model. Therefore, theoretically, even if the maximum value or the entire cross-section is not obtained, GA-IPPF will also achieve optimal solution exactly. To verify this view, we performed a set of simulation experiments to compare the final inversion results based on fully- coverage and half-coverage in each cross section.

In this section, we simulated spatial distributions of atmospheric CH<sub>4</sub> enhancements due to emissions of a strong point source. The configuration of simulations is as follow. Uncertainty of wind speed is 0.3 m/s, and uncertainty of wind direction is 30°. The concentration accuracy of sampling is 0.5 %. Other parameters are showed in Table.R1.

Table.R1. Parameters and retrieved results by two modes of sampling

Parameters	Actual	Full coverage (Fig.1a)	Half coverage (Fig.1 b)
Wind speed (m/s)	2.5	2.47	2.45
Wind direction (°)	90	90.1	89.7
Emission rate (kg/s)	300	298.3	304.1
a	0.11	0.11	0.104
b	0.93	0.92	0.931
c	0.1	0.099	0.103
d	0.93	0.925	0.933
H (m)	20	20.2	20.7
B (ppb)	2000	2002.3	2003.2

As is shown in Fig.R1 and Table R1, the retrieved emission rate bias is 0.6 % to the set emission of 300 kg/s in full coverage mode, while 0.13 % to 300 kg/s in half coverage mode, therefore, we regard sampling mode in Figure 3a has little impact on the final result.

If the mass balance method is used to quantify methane emission, the incomplete sampling of hot spots in a cross-section will definitely affect the final retrieved emission rate. The incomplete sampling of hot spots means that the number of molecules in the selected cross-section is smaller than the actual one, resulting in the underestimate of the final quantitative results. The unknown sampled concentration is indeed a major drawback during the flight. If the mass balance method is used in actual experiment, the long-distance sampling shall be conducted in the direction perpendicular to the wind direction of the cross section as far as possible to ensure the integrity of the hot spot collection.

The two hot spots you nonmentioned in Figure 3b, is caused by the phenomenon that according to Gaussian diffusion, at a certain height, the closer the spatial position to the downwind direction, the greater the diffusion value. We also carried simulation experiments to show the vertical distribution at different heights. Fig.R2 showed that there are different hot spots in different heights, and Fig 3b in our manuscript shows the diffusion of CH<sub>4</sub> in heights of 16 m, 25 m and 40 m, and some samples of UAV-based Aircore system are collected closer to the downwind direction, hence, they show higher values in each altitude.

For “the main lofted plume not resolved to the West at all”, it can refer to Fig.R1 and Table.R1. GA-IPPF would retrieve the unknown parameters according to different positions and the corresponding diffusion concentration. Half coverage mode would also ensure the high accuracy of the retrieved emission rate, see Table R1, the retrieved emission rate.

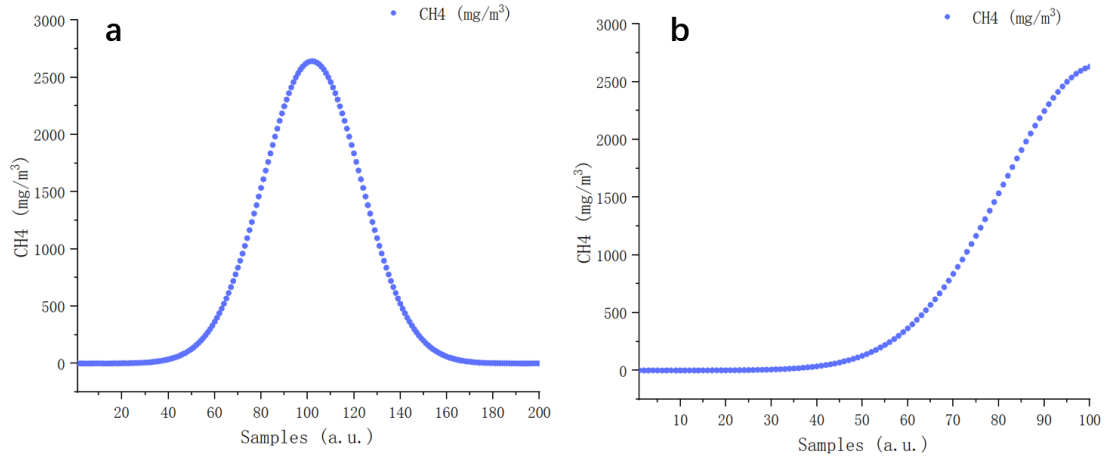


Fig.R1 Samples of two modes; a. full coverage; b. half coverage

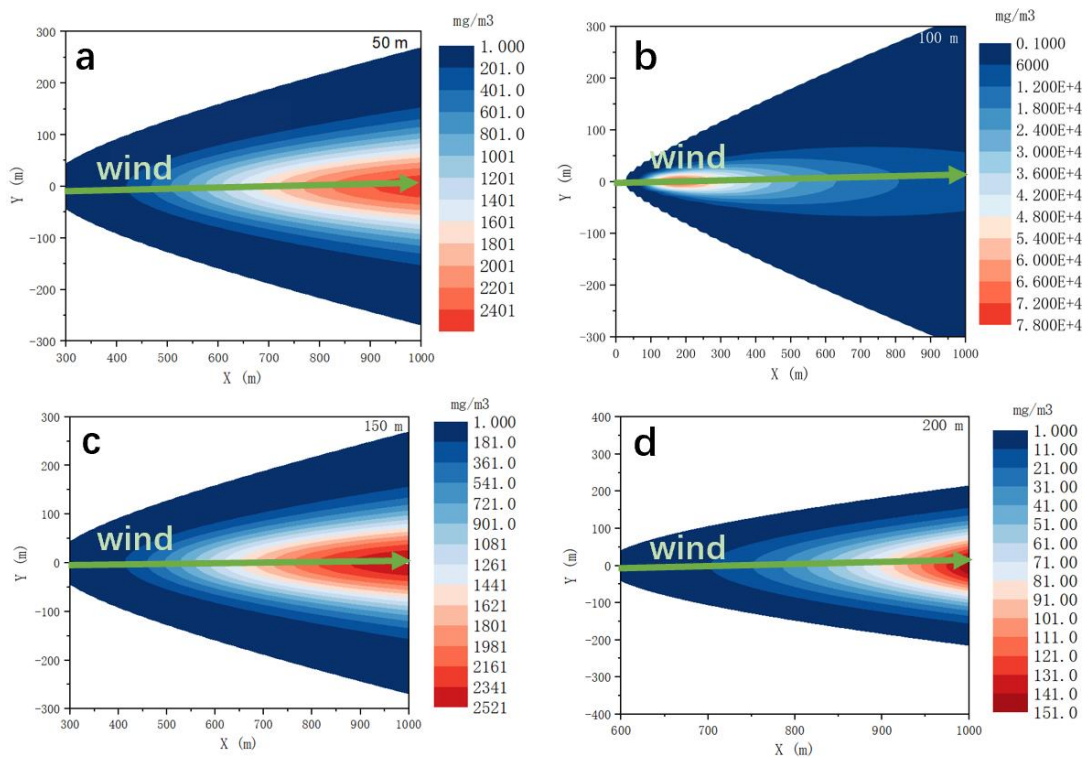


Fig.R2 Simulated diffusions of CH<sub>4</sub> at different heights, a.50 m; b.100 m; c.150 m and d. 200 m.

As for the real experiments, we add three release-controlled experiments as verification. The three experiments are UAV-based, vehicle-mounted and in-situ net respectively. The emission sources are spatially sampled, and then the emission intensity is retrieved by the quantitative GA-IPPF. Among them, the emission deviation between the intensity obtained by GA-IPPF and the actual release value are 3.2 %, 5.0 % and 3.2%, respectively, which proved a good quantitative effect.

Detailed responses in our manuscript are as follows:

### “Emission Estimates in control release experiment

To evaluate the performance of GA-IPPF in controlled release experiments, we quantified the emission rates in release experiment through different gases sample systems, including UAV-based Aircore system, mobile sampling system and ground-based in-situ network. Detailed introduction of the concerned release experiment are as follows:

#### Agrar Hauser control release

This CH<sub>4</sub> release experiment was conducted on Agrar Hauser field near Dübendorf, Switzerland. The controlled methane source was release from a 50 L high-pressure cylinder, the height of this artificial source is 1.5 m. meteorological information were acquired by 3D anemometers around the emission source. UAV-based sample systems used in these release experiments contains two sensors, including Quantum cascade laser spectrometer (QCLAS) and active Aircore. It carried a series active measurements from 23 February to 14 March 2020. There is no other CH<sub>4</sub> source around Agrar Hauser field and the topography is flat. In this section, active Aircore CH<sub>4</sub> samples on 12 March 2020 (312\_01) are chosen to use GA-IPPF to quantify methane release rate.

#### EPA methane control release

Environmental Protection Agency (EPA), USA developed OTM 33A method to quantify oil and gas leakage based on mobile measurement platforms, which consist CH<sub>4</sub> in-situ sensor (G1301-fc cavity ring-down spectrometer (Picarro)), a collocated compact weather station and a Hemisphere Crescent R100 Series GPS system. The accuracy of in-situ sample is within  $\pm 5\%$ , and in-situ sensor was implemented at height of 2.7 m based on vehicle. Weather station provides atmospheric temperature, pressure and humidity, as well as 3-D wind direction and wind speed. Simulated CH<sub>4</sub> leakage source was conducted using 99.9% methane high pressure cylinders as the gas supply. Total 20 experiments of control releases were published by EPA to evaluated OTM 33A.

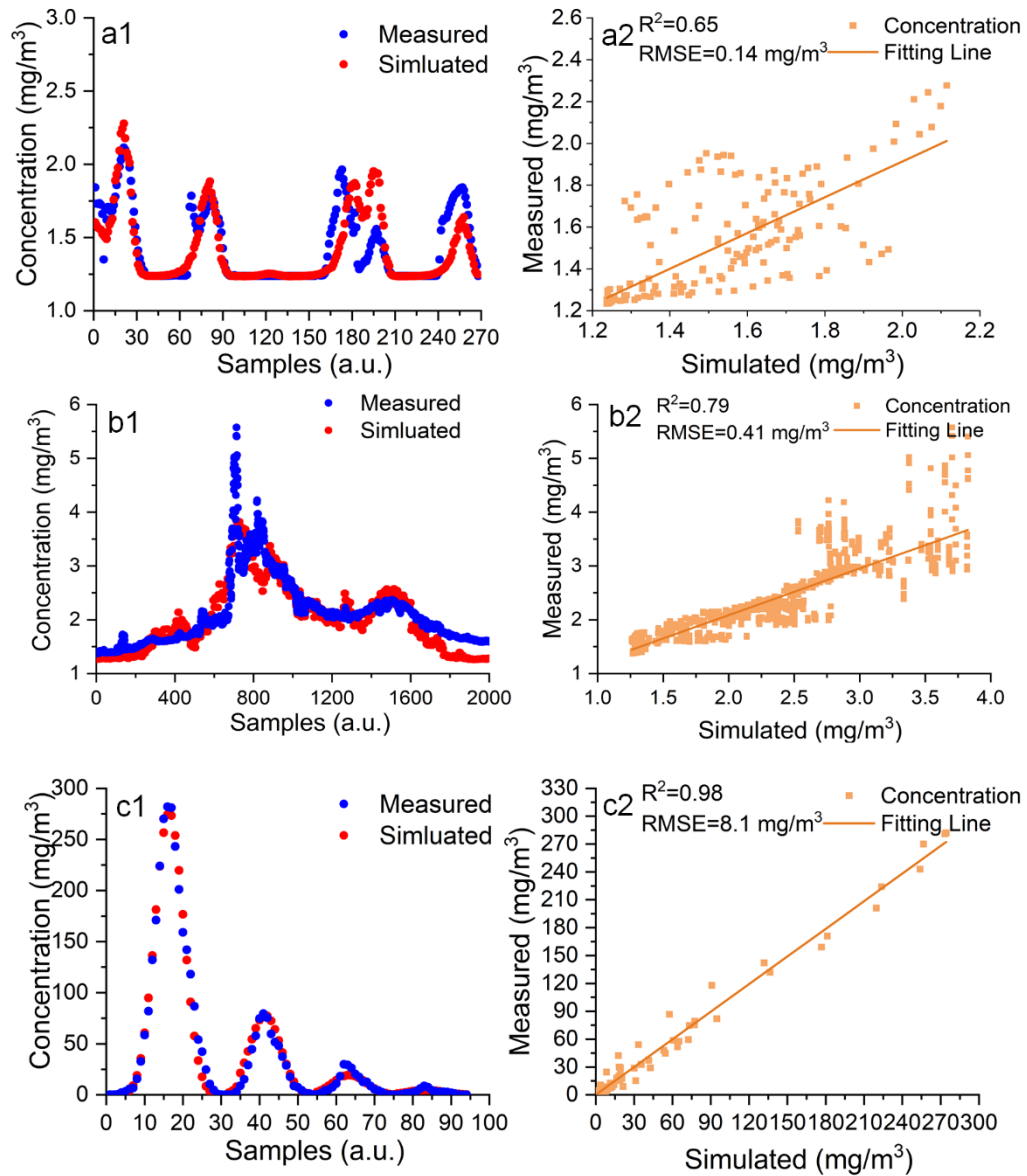
#### Prairie Grass emission experiment

Prairie Grass emission experiment was mainly to evaluate the diffusion of SO<sub>2</sub> from point source under different meteorological circumstances [Barad, 1958], the height of emission source is 0.46 m, all in-situ sensors are set at heights of 1.5 m. SO<sub>2</sub> concentration sampled by in-situ network at radius of 50 m, 100 m, 200 m, 400 m and 800 m. Samples in R57 release (10-minute sampling periods), total 94, were selected to quantified SO<sub>2</sub> emission rate from release instrument. Reported emission rate of SO<sub>2</sub> in R57 is 101.5 g/s, samples collected in radius of 800 m was neglected in this discussion for amount of what is extreme less. Reported wind speed is  $4.85 \pm 1$  m/s, wind direction is  $184 \pm 10^\circ$ .

**Table 4 Performance of GA-IPPF model in different control release experiments**

Database	Number	Gas	Release rates (g/s)	Retrieved by GA-IPPF (g/s)
Agrar Hauser	312_01	CH <sub>4</sub>	$0.31 \pm 0.03$	$0.3 \pm 0.03$
EPA	STR_6061411_01	CH <sub>4</sub>	0.60	$0.57 \pm 0.04$
Prairie Grass	57	SO <sub>2</sub>	101.5	$104.7 \pm 3.7$

Table 4 shows the emission rates and uncertainties through GA-IPPF in control release experiments, and the reported emission rates. The average difference between retrieved emission rates and reported ones is 3.8 %, which indicates the high accuracy of GA-IPPF in quantification estimation.



**Fig.10.** Rebuild the gases diffusion based on retrieved parameters in control release experiments; a1 and a2 are comparisons between simulated diffusion and actual samples in Agrar Hauser; b1 and b2 are comparisons between simulated diffusion and actual samples in EPA control release; c1 and c2 are comparisons between simulated diffusion and actual samples in Prairie Grass experiment.

As is shown in Fig.10, the rebuild diffusions of gases by GA-IPPF in three kinds of control release experiment are logical, consistencies of simulated gases concentration and actual samples are good(see Fig.10.a1, Fig.10.b1 and Fig.10.c1), each peaks of the samples in control release experiment could be reconstructed. The correlations between simulated gases concentration and actual samples are larger than 0.65, and the RMSE are within 2.7% (relative to the mean value of the selected samples' concentration). In general, reconstruction of gases concentration in both mobile-platform and UAV-based data are worse than that in in-situ network. Collected data in in-situ network are usually the mean value of a certain time, like 10 min in Prairie Grass emission experiment, which provide stabile inputs data in GA-IPPF, especially concentration samples. While the concentration samples in mobile-platform and UAV-based Aircore experiments is instantaneous, which may be inaccurate and exist fluctuations in collections. The advantages of vehicle-based and UAV-based sample systems are flexibility, which could acquire the

distribution of gases around the target monitoring sources freely. In-situ network is complicated to implement and the cost is high, and the wind direction should be considered when deploying. But its high stability and accuracy could help us to quantify emission source. Therefore, environmental protection departments can choose detection systems according to actual emission monitoring needs.

”

2. The issue of quantification of methane from the energy sector has the potential to become potentially political and legal – with the concept of emissions levies or preferred contracts based on quantified methane emissions being used as a legal instrument. It is therefore of utmost importance that the authors of novel methodologies for quantification such as those shown here are aware of how their methods may be utilised in the future, potentially by 3rd party commercial companies, and ensure that any issues are fully declared (e.g. why are only 2 flights used to verify the method, were there occasions when this method failed, and why? There are 15 flights stated on the experiments section). They should also check that all references to accuracy are truly discussing accuracy (closeness to a true value) – and not precision (closeness to another measurement or model).

Thank you for your useful suggestion, it's critical to note that the requirement of the collected data by different measured instruments in GA-IPPF. Because the national government may develop different sample systems based on different platforms to monitor the methane emission. If GA-IPPF become a criterion in government monitoring policy, the limitation and application of GA-IPPF should also be introduced in this work to validate the reliable.

In the latest revised version, we expounded the matters needing attention and process of GA-IPPF for users, which as follows:

1. Meteorological instruments should be equipped during the concentration sampling, which could acquire wind speed, wind direction, humidity and atmospheric pressure.
2. The wind speed should not be too low, and we recommended it should be larger than 2m/s.
3. During actual experiments, after the stable wind speed and wind direction were measured, the UAV-based Aircore system should try to fly along the cross section perpendicular to the wind direction.

Actually, we totally collected 15 Flights data around Pniówek coal on 18/08/2017 and 21/08/2017. Therefore, we initially considered choosing one Flight to discuss mine methane emissions detailly using the GA-IPPF model as an example.

In this work, the Flights meet the two criterions has been selected in the analysis of methane quantification. Firstly, the wind speed is larger than 2 m/s, which would possible lead stable wind direction. Secondly, the flight trajectory of UAV is nearly perpendicular to the wind direction (within  $15^\circ$  ). Finally, eight Flights fulfilled the set requirements simultaneously.

As you mentioned that we should compare the emission rate calculated by GA-IPPF with actual known Pniówek coal emission rate in same time. It's really reasonable, however, we didn't know actual instantaneously methane emission of Pniówek coal during the Aircore Flight. Therefore, we prove the accuracy of GA-IPPF through indirect comparison, mainly including two parts:

1. Comparison with other quantification methods, including emission-factories, NLSL, IPPF as well as mass balance and Gaussian inversion. The results calculated by different methods show differences, and the results calculated by GA-IPPF and these evaluation methods are in the same

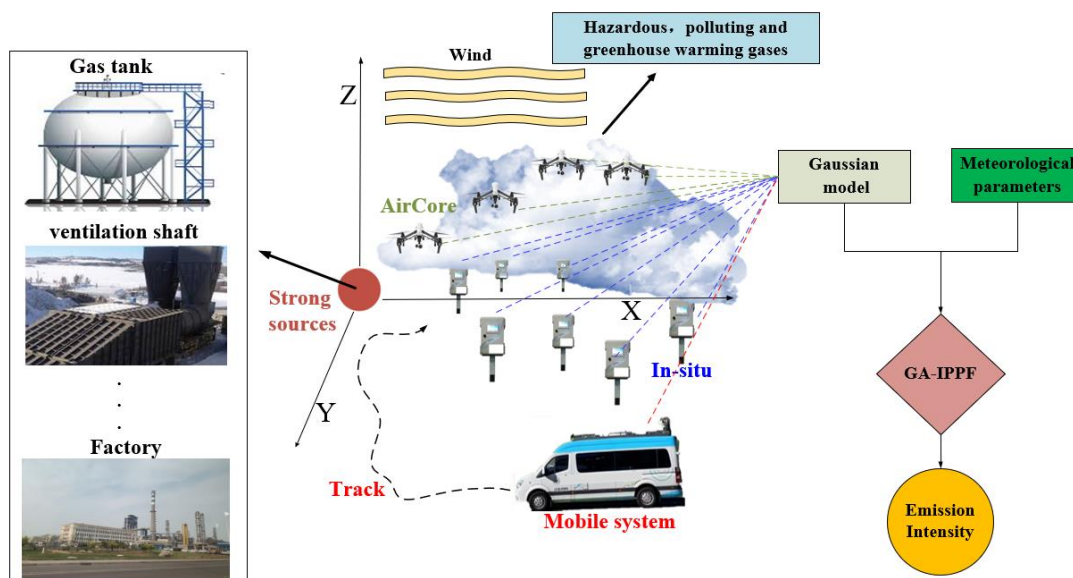
order of magnitude among the retrieved emission rate, and between the maximum and minimum values of retrieved emission rates.

2. We evaluated the accuracy of GA-IPPF in three release-controlled experiments, which could evaluate the performance of GA-IPPF when actual emission rates are known, and the bias between retrieved emission rates and reported ones within 5.0 %, see table 4.

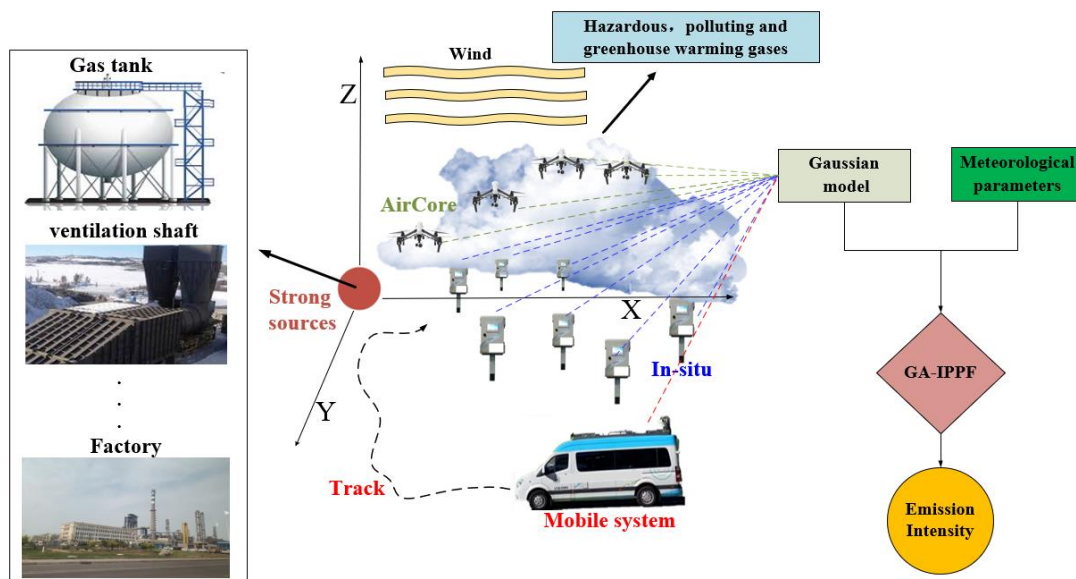
**Table 4 Performance of GA-IPPF model in different control release experiments**

Database	Number	Gas	Release rates (g/s)	Retrieved by GA-IPPF (g/s)
Agrar Hauser	312_01	CH <sub>4</sub>	0.31±0.03	0.3±0.03
EPA	STR_6061411_01	CH <sub>4</sub>	0.60	0.57±0.04
Prairie Grass	57	SO <sub>2</sub>	101.5	104.7±3.7

3.If this method is suitable, I think a comment section needs to be added discussing other sorts of data that the team would expect this method to work with. I'm assuming that on-board drone measurements downwind from landfill / industrial sites would be a good option, would mobile measurements from vehicles potentially work?



Thank you for this suggestion. According to the characteristic of GA-IPPF model, it can be seen that the input data mainly includes location information of the samples, the concentration of the samples and the meteorological parameters. As long as the researchers or the energy monitoring department can obtain these collection data, GA-IPPF can be applied to quantify gases emission of industrial sites, gas leak from gas tank, etc.



**Fig.9.** Application of GA-IPPF in quantifying emission source of gases through different sample systems ; including UAV-based Aircore system, ground-based In-situ network and mobile collection system.

In order to better demonstrate the applicability of this method, we have added a chapter named “ Application of GA-IPPF”, showing the performance of GA-IPPF in three different release-controlled experiments, as follows:

As for the real experiments, we added three release-controlled experiments as verification. The three experiments are UAV-based, vehicle-mounted and in-situ net respectively. The emission sources are spatially sampled, and then the emission intensity is retrieved by the quantitative GA-IPPF. Among them, the emission deviation between the intensity obtained by GA-IPPF and the actual release value are 3.2 %, 5.0 % and 3.2%, respectively, which proved a good quantitative effect.

Detailed responses in our manuscript are as follows:

#### “Emission Estimates in control release experiment

To evaluate the performance of GA-IPPF in controlled release experiments, we quantified the emission rates in release experiment through different gases sample systems, including UAV-based Aircore system, mobile sampling system and ground-based in-situ network. Detailed introduction of the concerned release experiment are as follows:

#### **Agrar Hauser control release**

This CH<sub>4</sub> release experiment was conducted on Agrar Hauser field near Dübendorf, Switzerland. The controlled methane source was release from a 50 L high-pressure cylinder, the height of this artificial source is 1.5 m. meteorological information were acquired by 3D anemometers around the emission source. UAV-based sample systems used in these release experiments contains two sensors, including Quantum cascade laser spectrometer (QCLAS) and active Aircore. It carried a series active measurements from 23 February to 14 March 2020. There is no other CH<sub>4</sub> source around Agrar Hauser field and the topography is flat. In this section, active Aircore CH<sub>4</sub> samples on 12 march 2020 (312\_01) are chosen to use GA-IPPF to quantify methane release rate.

#### **EPA methane control release**

Environmental Protection Agency (EPA),USA developed OTM 33A method to quantify oil and gas leakage based on mobile measurement platforms, which consist CH<sub>4</sub> in-situ sensor (G1301-fc cavity ring-down spectrometer (Picarro)), a collocated compact weather station and a Hemisphere



Crescent R100 Series GPS system. The accuracy of in-situ sample is within  $\pm 5\%$ , and in-situ sensor was implemented at height of 2.7 m based on vehicle. Weather station provides atmospheric temperature, pressure and humidity, as well as 3-D wind direction and wind speed. Simulated CH<sub>4</sub> leakage source was conducted using 99.9% methane high pressure cylinders as the gas supply. Total 20 experiments of control releases were published by EPA to evaluate OTM 33A.

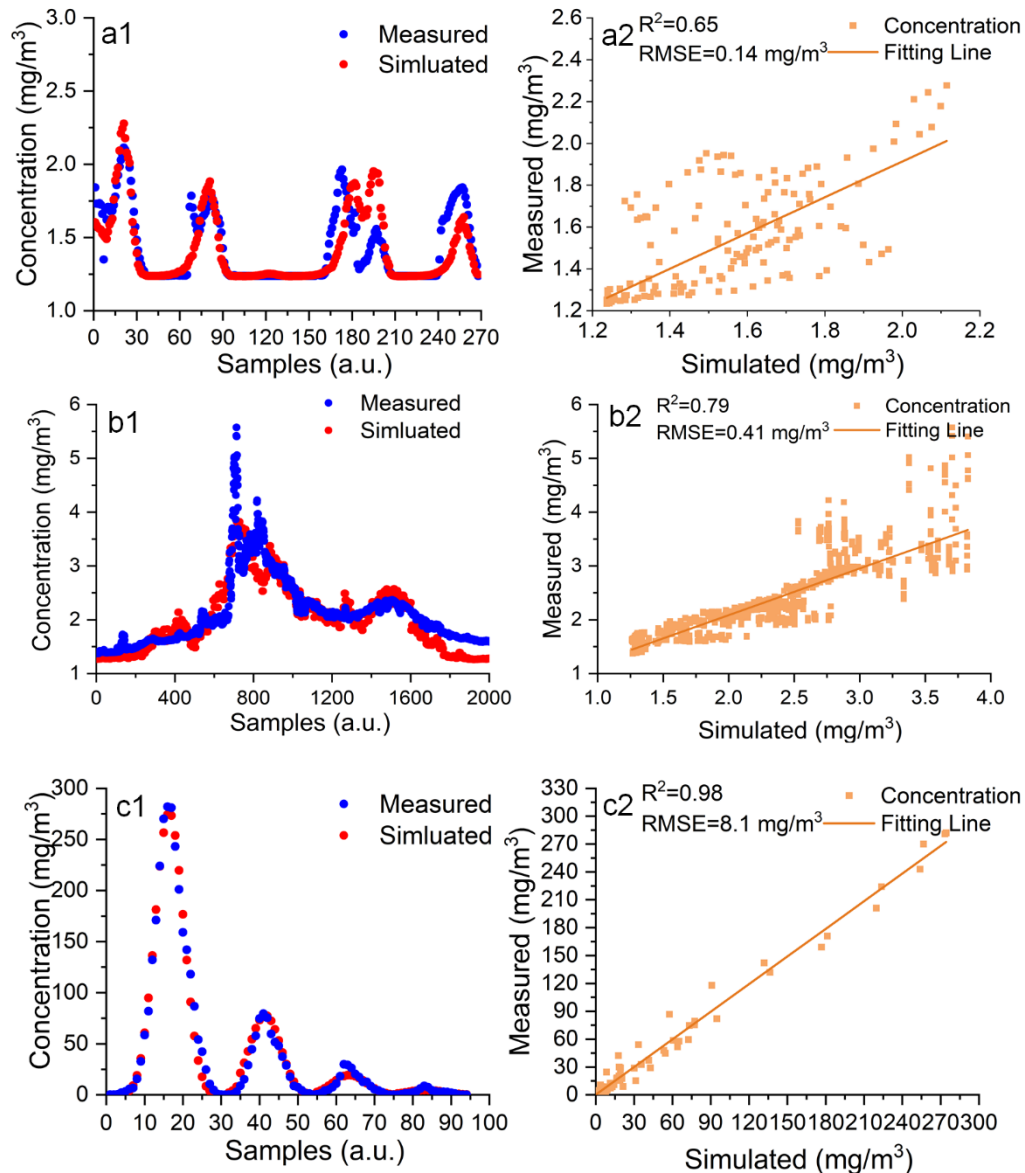
### **Prairie Grass emission experiment**

Prairie Grass emission experiment was mainly to evaluate the diffusion of SO<sub>2</sub> from point source under different meteorological circumstances [Barad, 1958], the height of emission source is 0.46 m, all in-situ sensors are set at heights of 1.5 m. SO<sub>2</sub> concentration sampled by in-situ network at radius of 50 m, 100 m, 200 m, 400 m and 800 m. Samples in R57 release (10-minute sampling periods), total 94, were selected to quantify SO<sub>2</sub> emission rate from release instrument. Reported emission rate of SO<sub>2</sub> in R57 is 105.1 g/s, samples collected in radius of 800 m were neglected in this discussion for amount of what is extremely less. Reported wind speed is  $4.85 \pm 1$  m/s, wind direction is  $184 \pm 10^\circ$ .

**Table 4 Performance of GA-IPPF model in different control release experiments**

Database	Number	Gas	Release rates	Retrieved by GA-IPPF
Agrar Hauser	312_01	CH <sub>4</sub>	$0.31 \pm 0.03$ g/s	0.3
EPA	STR_6061411_01	CH <sub>4</sub>	0.60 g/s	0.57
Prairie Grass	57	SO <sub>2</sub>	101.5 g/s	104.7

Table 4 shows the emission rates and uncertainties through GA-IPPF in control release experiments, and the reported emission rates. The average difference between retrieved emission rates and reported ones is 3.8 %, which indicates the high accuracy of GA-IPPF in quantification estimation.



**Fig.10.** Rebuild the gases diffusion based on retrieved parameters in control release experiments; a1 and a2 are comparisons between simulated diffusion and actual samples in Agrar Hauser; b1 and b2 are comparisons between simulated diffusion and actual samples in EPA control release; c1 and c2 are comparisons between simulated diffusion and actual samples in Prairie Grass experiment.

As is shown in Fig.10, the rebuild diffusions of gases by GA-IPPF in three kinds of control release experiment are logical, consistencies of simulated gases concentration and actual samples are good(see Fig.10.a1, Fig.10.b1 and Fig.10.c1), each peaks of the samples in control release experiment could be reconstructed. The correlations between simulated gases concentration and actual samples are larger than 0.65, and the RMSE are within 2.7% (relative to the mean value of the selected samples' concentration). In general, reconstruction of gases concentration in both mobile-platform and UAV-based data are worse than that in in-situ network. Collected data in in-situ network are usually the mean value of a certain time, like 10 min in Prairie Grass emission experiment, which provide stabile inputs data in GA-IPPF, especially concentration samples. While the concentration samples in mobile-platform and UAV-based Aircore experiments is instantaneous, which may be inaccurate and exist fluctuations in collections. The advantages of vehicle-based and UAV-based sample systems are flexibility, which could acquire the

distribution of gases around the target monitoring sources freely. In-situ network is complicated to implement and the cost is high, and the wind direction should be considered when deploying. But its high stability and accuracy could help us to quantify emission source. Therefore, environmental protection departments can choose detection systems according to actual emission monitoring needs.

In summary, GA-IPPF could apply in different sample systems to quantify emission rate from strong sources with high flexibility.

### Minor

I am far from a professional proof-reader, and would recommend that this manuscript is looked over by a proofer after corrections as there are numerous tense and grammar issues that need resolving to make the paper read as desired.

Dear reviewer, we are sorry for our wrong tense and grammar in our original manuscript. And we have checked and modified these wrong parts in our latest manuscript, and we have inquired Copernicus Publications for the recommended Professional English editing company. They informed once our manuscript be accepted for final publication, they will perform an extensive copy-editing for English with the goal to ensure the paper adheres to scientific writing standards, grammatical accuracy, and overall readability.

**L20: There are plenty of monitoring methods – but very little verified quantification methods suitable for coal mines.**

Thank you for your suggestion on this sentence, we have modified this sentence to make the statement more reasonable as follows:

“There are plenty of monitoring methods to quantify gases emission rate based on gases concentration samples around the strong sources. However, there is a lack of quantitative models to evaluate methane emission rate from coal mines with less priori information.”

**L34: Grammar. Release is concerning.**

Thank you for your kindly comment on this sentence, we have modified this sentence as follows:

“The release of CH<sub>4</sub> into the atmosphere during coal mining is very concerning because it contributes to increased atmospheric concentration of CH<sub>4</sub>, one of the most important greenhouse gases and is a waste of resources.”

**L38: Why are BU only useful for strong sources?**

We apologize for this wrong expression, actually, besides strong emission sources, BU can provide also the emission rate or gases flux of grids with different spatial resolution. And we has modified this sentence as follows:

“Bottom-up inventories can provide us with CH<sub>4</sub> emission rates from strong point sources or gridded CH<sub>4</sub> fluxes with different spatial resolutions, which play a great role in statistical analysis.”

**L42: This seems to be a common misuse / expectation of a BU inventory. They are not intended to be able to capture variable emissions, but are a statistical average expectation. It is only worth considering inventories compared to spot measurements if there is some valid statistical analysis (either temporal, or numbers of sites)**

We're sorry for this inappropriate expression. BU does play an important role in statistical analysis, such as comparison of different national emission rates, statistical basis for environmental protection policies and energy distribution in different industry sectors, et. al. We wrongly expressed negative appraisal on its function in real-time emissions evaluation.

Therefore, we have modified the original text as follows:

“Bottom-up inventories can provide us with CH<sub>4</sub> emission rates from strong point sources or gridded CH<sub>4</sub> fluxes with different spatial resolutions, which play a great role in statistical analysis. However, the low temporal resolution of inventory data does not allow us to obtain emission intensity from target sources instantaneously.”

**L44: What improvements have suddenly made this possible?**

Sorry for this incomplete expression, we have modified this sentence as follows:

“With the development of different atmospheric CH<sub>4</sub> concentration measurement techniques, like Fourier spectrometer, differential absorption Lidar, Aircore system, and in-situ sensors, CH<sub>4</sub> emission rates for strong emission sources could be quickly quantified by top-down methods with high accuracy.”

**L46: Tense: should be is capable of obtaining.**

Thank you, and we have revised this sentence as follows:

“Greenhouse gases observing satellite (GOSAT) and TROPOspheric Monitoring Instrument (TROPOMI) are capable of obtaining the column concentration of CH<sub>4</sub> (XCH<sub>4</sub>, ppb) with spatial resolution of 10 km×10 km and 5 km×7.5 km respectively.”

**L59: But most aircraft equipped with any CH<sub>4</sub> sensor are able to achieve ppb precision and are perfectly capable of measuring downwind flight plans from coal mines. Many aircraft are capable of this such as Scientific Aviation et.**

We are sorry for this wrong comment on the aircraft remote sensing systems in our original manuscript. It's correct that most aircraft equipped with CH<sub>4</sub> sensors are able to achieve ppb precision and are perfectly capable of measuring downwind CH<sub>4</sub> of coal mines. Such as Methane Airborne MAPper (MAMAP) instrument(Krautwurst et al., 2021), and airborne-based AVIRIS-NG system, both of them could acquire the concentration in downwind direction with reasonable flight track. We have modified sentence in Line 59 as follows:

“An airborne vehicle could fly at low altitudes to improve the acquisition of CH<sub>4</sub> concentration (Elder et al., 2020; Wolff et al., 2021; Krautwurst et al., 2021) and estimate CH<sub>4</sub> emission from strong sources by the cross-sectional flux method or the Gaussian dispersion method. However, the cost of airborne experiment is high and the fly plan is easily to be restricted by aviation control policies.”

**L62: What about ground based eddy covariance?**

We are sorry for neglecting the introduction for ground-based eddy covariance, and we have added this information in our latest manuscript as follows:

“Ground-based eddy covariance sites could monitor agriculture and forestry ecology methane flux with high temporal resolution(Jha et al., 2014), such as mangrove ecosystem, larch forest in eastern Siberia(Nakai et al., 2020).”

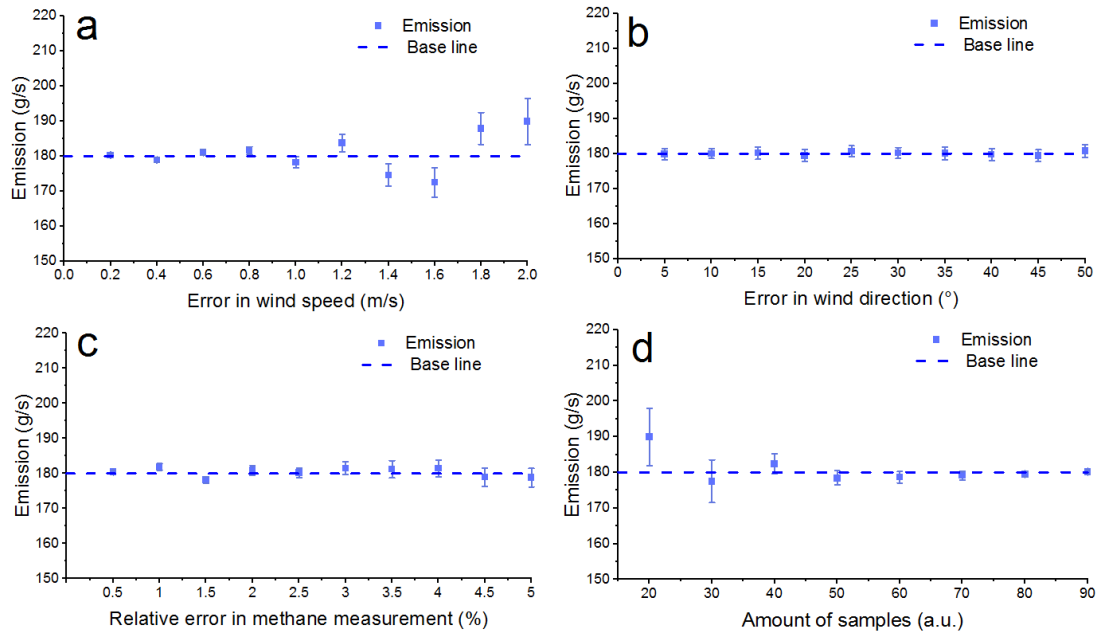
**General: to L70. This section feels unnecessarily negative about the capabilities of the rest of the scientific community with regards to being able to measure emissions from coal mines. There are several methods discussed here that I would envisage perfectly capable of making precise measurements that could enable a quantification of emission estimate.**

Sorry for the unnecessarily negative comments we made on different detection technologies in the original manuscript. In the latest submission, we have introduced the application, advantages and disadvantages of each detection technology in quantifying methane emissions. As we know, any detection technique has its own unique advantages and limitations. We have re-edited this paragraph in latest submission as follows:

“Greenhouse gases observing satellite (GOSAT) and TROPOspheric Monitoring Instrument (TROPOMI) are capable of obtaining the column concentration of CH<sub>4</sub> (XCH<sub>4</sub>, ppb) with spatial resolution of 10 km×10 km and 5 km×7.5 km respectively. The regional CH<sub>4</sub> flux can be retrieved by assimilating the measured XCH<sub>4</sub> into an atmospheric dispersion model (Tu et al., 2022; Feng et al., 2016). PRISMA hyperspectral imaging satellite and GHGsat can detect increased CH<sub>4</sub> caused by strong emission sources with high spatial resolutions, and the comprehensive CH<sub>4</sub> emission can be quantified by integrated mass enhancement or cross-sectional flux method (Guanter et al., 2021; Varon et al., 2020). It plays a huge role in the analyzing methane emission rate from strong sources, but it has high requirements for satellites’ detection track, that is, to monitor the methane distribution in the target area within coverage range (Schneising et al., 2020; Varon et al., 2019). Airborne sensors can fly at low altitudes to improve the acquisition of CH<sub>4</sub> concentration data and estimate CH<sub>4</sub> emission from strong sources by the cross-sectional flux method or the Gaussian dispersion method (Elder et al., 2020; Wolff et al., 2021; Krautwurst et al., 2021). It enables repeated monitoring of emission sources in a large area in a short period of time, however, airborne experiments’ cost is high and the flight tracks may be restricted by the aviation control policies. Ground-based eddy covariance sites can monitor agriculture and forestry ecology methane flux with high temporal resolution, such as mangrove ecosystem (Jha et al., 2014), larch forest in eastern Siberia (Nakai et al., 2020). Its accuracy is very high, but there is currently less monitoring of methane emissions from strong point sources using eddy covariance. When ground-based concentration sensors fixed in appropriate position, they have the advantage of continuously sampling gas concentration in downwind direction from the source. It will provide important dispersion data for methane emission quantification model at the enterprise level, but these sensors usually need to be carried on a vehicle platform to obtain methane concentration at different locations (Zhou et al., 2021; Robertson et al., 2017; Caulton et al., 2017). Ground-based differential absorption LIDAR can obtain the CH<sub>4</sub> profile concentration in different altitudes, whose data is suitable as the input of the emission-retrieval model (Shi et al., 2020a), but it has high requirements in terms of hardware performance and system stability (Shi et al., 2020b). An unmanned aerial vehicle (UAV) can reach any location rapidly around the CH<sub>4</sub> sources, which can sample CH<sub>4</sub> concentration with location information (Nathan et al., 2015; Iwaszenko et al., 2021), when equipped with concentration sensors. It can acquire the distribution characteristics when sufficient concentration data are collected, which is beneficial to retrieving emission rate. The cost of UAV-based AirCore system is low and the process of its sample data is relatively simple, but the diffusion of methane emitted from strong sources may be sampled incompletely.”

**L73: What does high applicability mean in this sense?**

Sorry for the unclear expressing here. Actually, what we want to express is that the most outstanding advantage of Aircore system is collecting methane concentration at different locations freely, thus it can obtain useful data to rebuild emission diffusion, contains spatial location and corresponding concentration. Then, GA-IPPF have a self-adjusting effect on the influence of errors in detection accuracy and the acquisition of meteorological conditions. See the figure below for details, all of which have the effect of reducing errors.



**Fig. 8.** Influence of accuracy of parameters on retrieved emission results. The baseline represents the emission rate setting of CH<sub>4</sub>, 300 g/s: (a) wind speed, with additional error ranging within 0.2–2 m/s and an interval of 0.1 m/s, (b) wind direction, with additional error ranging within 5°–50° and an interval of 5 °, (c) accuracy of CH<sub>4</sub> samples, with additional error ranging within 0.5%–5.0% and an interval of 0.5%, and (d) amount of CH<sub>4</sub> samples, randomly selected as 20–90 among the defined 99 samples.

We have revised this sentence as follows :

“In 2017, we developed an UAV-based active AirCore system, which could sample spatial atmospheric CO<sub>2</sub>, CH<sub>4</sub>, and CO with high accuracy (Andersen et al., 2018), aiming to retrieve greenhouse gases emission of strong sources. The most urgent issue we need to address is to develop an emission quantification model to make use of the advantage of the collected data by Aircore, namely collecting data at different locations with high flexibility.”

L74: How will it have less uncertainty if the inputs still have the same uncertainties attached to the actual measurements? The discussion to L83 makes it seem that these important atmospheric parameters can be discarded in favour of a set of model parameters? From experience at controlled release experiments small changes in wind direction and other meteorological conditions can have a dramatic effect on the plume behaviour.

Dear reviewer, this question you raised is very meaningful. According to the formulations of the Gaussian Diffusion Model, small changes in wind direction and speed, as well as other meteorological conditions, can indeed cause significant impact on the final retrieved emission results.

The original meaning of sentence (line L83), which also highlights the shortcoming of the existed methods, once there are errors in meteorological data, it will cause great impact on the final assessment. In Fig 8, we discussed the influence of different uncertainty of GA-IPPF in wind speed and wind direction on the final calculation through simulations. It worth nothing that during the process of optimization, the uncertainty of the original input of wind speed and wind direction has been added. For example, wind speed was fluctuated within  $\pm 2$ m/s and wind direction was fluctuated within  $\pm 60^\circ$ . During the process of GA-IPPF, upper and lower limits are set for wind

speed and wind direction. For example, if the original wind speed is 3m/s, original wind direction is 80°, the lower limit of wind speed is 1m/s, and the upper limit is 5m /s, the lower limit of the wind direction is 20°, and the upper limit is 140°. The wind speed will have a linear relationship with the final diffusion; the wind direction determines the location of the sampling point, and the concentration of sampling points in different locations represent the characteristic of diffusion from emission source. Because GA-IPPF will calculate the global optimal solution for all the unknown parameters, so important parameters such as wind speed and wind direction will be dynamically adjusted within lower and upper limits. The objective function we set is served as the judgment of GA-IPPF, that is, the reconstructed data is consistent with the real sampled data. Through the optimal adjustment of GA-IPPF, the objective function would meet its minimum value. In actual measurement, these meteorological parameters cannot be discarded in theory. However, in the case of insufficient funds or technology, some researchers or energy departments have not installed meteorological instruments in field experiments. In this situation, the alternatives are given in section 3.3, and we recommend meteorological reanalysis data to participate in GA-IPPF. Through comparison, it is found that the deviation between the emission intensity obtained by using meteorological data and meteorological analysis data is within 20%. We believe that once the limits of each parameter are set appropriately, the accuracy of inversion can achieve satisfactory results. The advantage of the GA-IPPF model is that all unknown parameters in Gaussian dispersion model are dynamically adjusted, reducing the accuracy requirements of the collected data in actual experiments.

**L106: Is the accuracy only 20ppb using a G2401-m? This is concerning, why is it so large? What is the location accuracy of the sampling, how much does the sample bleed into itself over the course of a flight? Is the mixing in the Aircore linear across flight time?**

Indeed, the precision of methane measurements was 20 ppb, because we used a high-range CH<sub>4</sub> mode of the CRDS, and the normal mode would not be able to measure up to 125 ppm. This is actually not an issue as the relative uncertainty of 20 ppb with respect to 100 ppm is only 0.02%. The spatial resolution of the AirCore measurements is estimated to be 18-40 m with a UAV flight speed of 1-2 m/s. Since the spatial resolution is dominated by the sample smearing in the cavity of the CRDS analyzer and the air samples were analyzed quickly after landing, the spatial resolution was only affected by the UAV flight speed.

**L117: correlation not connection? How good a correlation?**

Dear reviewer, thank you for this suggestion, we are sorry for we haven't expressed this sentence clearly. We have modified this sentence in latest manuscript as follows:

“During the detection, the spatial position of the radiosonde is well recorded by satellite, we assume that the uncertainty in the wind direction is low.”

**L135: I'm not 100% sure what alpha represents, please clarify (with a reference if possible)**

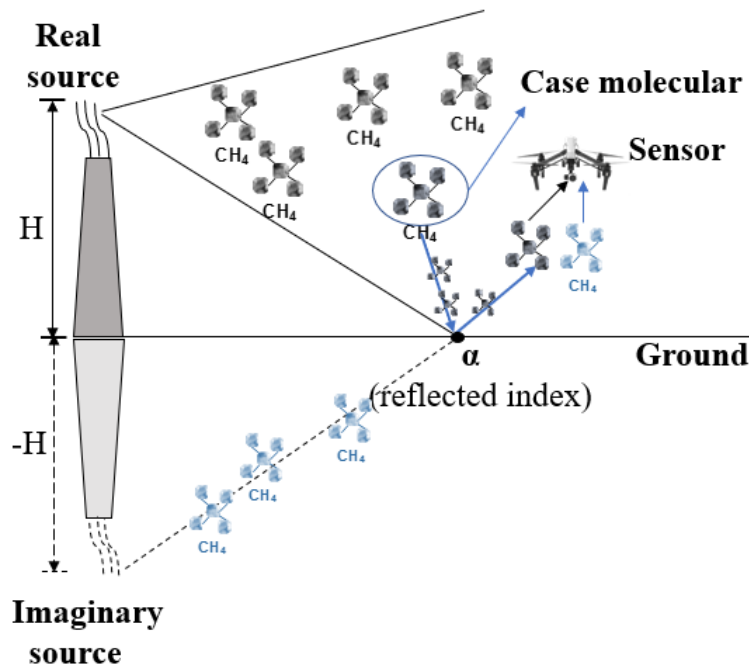


Fig R3. Introduction to  $\alpha$  in Gaussian dispersion model

Dear reviewer,  $\alpha$  is the reflected index of the diffusion gases when the gas molecular touch the ground, it can be simply treated as the ejection effect. For example, if the gas molecular catapult into the atmosphere totally,  $\alpha = 1$ . Conversely, if the gas molecular absorb by ground totally, no ejection effect,  $\alpha = 0$ .  $\alpha$  is relative with the actual field circumstance and the diffusion molecular.

Principle:

In Fig.R3, we take  $\text{CH}_4$  dispersion as an example.  $H$  is the effectively emission height of real source, assuming an imaginary source symmetrical to the ground. The imaginary is also regarded as a potential emission source, the sensor sample concentration of  $\text{CH}_4$  is the sum of  $a$  and  $\alpha \times b$ . therefore,  $\alpha$  would determine the value of samples in certain position. You could reference this item according to page 36 in <https://ansn.iaea.org/Common/Topics/OpenTopic.aspx?ID=13012>.

Approx. L160. Is there prioritization in the fitting process? Are some variables given priority due to their certainty? General question about the process, if the inputs are very close to the outputs, then I presume that the standard gaussian plume quantification would also be very close?

Dear reviewer, thank you for this comment. The concept similar to priority is existed in our proposed model, refers to the upper and lower bounds we set for each unknown parameter. For example, during the actual experiments, if uncertainty of wind speed is relatively small, we would set smaller range for its dynamic adjustment, which means we set larger weight constraints for wind speed, which is similar to the concept of priority of wind speed in final emission quantification.

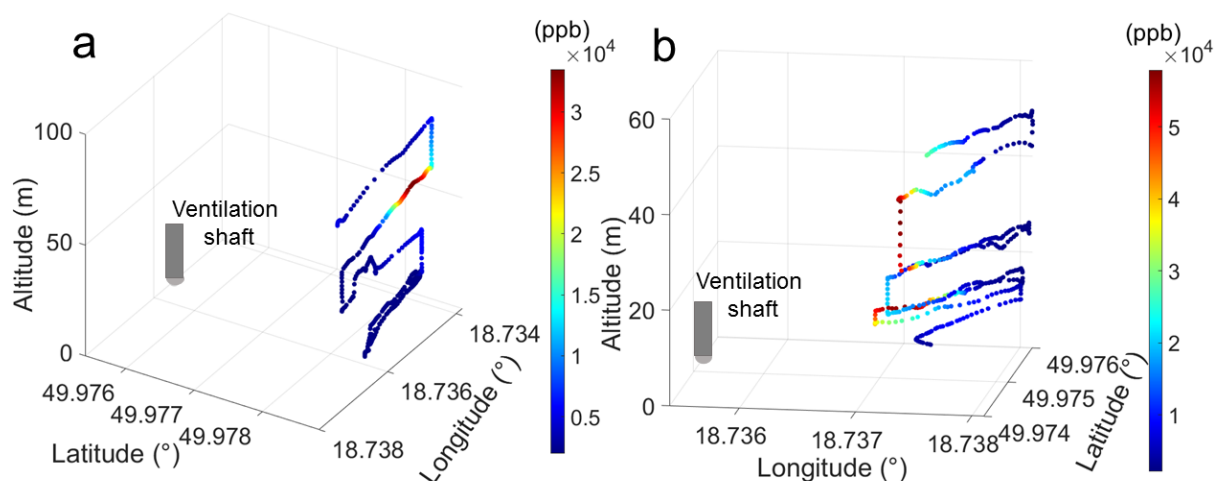
The relationship between value of inputs and output value cannot directly judge whether the Gaussian quantification is reliable. In GA-IPPF, input parameters in final step of emission calculating process are obtained according to the repeated fitting results by genetic algorithm. During the solution of IPPF, we set fluctuation for each parameter based on the calculated results by genetic algorithm. The final output values are retrieved according to the judgment condition we set, refers to the relationship between the rebuild diffusion calculated by retrieved parameters and actual samples. Therefore, we don't judge the accuracy of Gaussian plume quantification based on the consistency of the input and output values. In fact, RMSE and  $R^2$  between the reconstructed



diffusion concentration and the actual samples are worked as judgement, when the RMSE is small and R2 is high, that means, the Gaussian quantification for the emission sources is better.

L190: If these are spiral patterns, then the figure doesn't make this very clear. Can these be replotted to make it clear if that is the case?

The samples of Aircore is the concentration of methane in certain position, it is not a continuous data acquisition. The sample trajectory of UAV is nearly a cross-section on two-dimensional plane, which cannot show the samples with obvious 3d Visual. In latest submission, we tried our best to change the visual angle to make Fig 3 closer to 3D visual as follows:



Para 218: This paragraph needs tidying up, it is unclear as it currently reads and has numerous typos. The reconstruction is impressive – especially with two peaks in Flight 15 so it is important that this section is as simple and clear as possible.

Thank you for this suggestion, we have modified this paragraph in latest submission. And as you mentioned the two peaks in Figure 3 (b), it was accused by the diffusion characteristic from strong point sources, in order to make it easier to understand, we simulated the diffusion at different altitudes as follows:

Table R1. Parameters settings

Parameters	value
Wind speed (m/s)	2.5
Wind direction (°)	90
Emission rate (kg/s)	300
a	0.11
b	0.93
c	0.1
d	0.93
H (m)	20
B (ppb)	2000

We also carried simulation experiments to show vertical distribution at different heights. As is shown in Fig.R2, it would exist different hot spot in different height.

In Fig.R2, we could see diffusion of gas exist higher value in each height, for example, the diffusion of 50m and 150m are in same magnitude. Fig 3b is show the diffusion of CH<sub>4</sub> in height of

22.3 m and 52.4 m, the two spots are around the wind direction (X axis), thus, it exist two spots in Fig 3 b. If the UAV's Flight across this two heights, it would be possible that the samples exist two hots, which would be explained for the two peaks in Fig. 3(b).

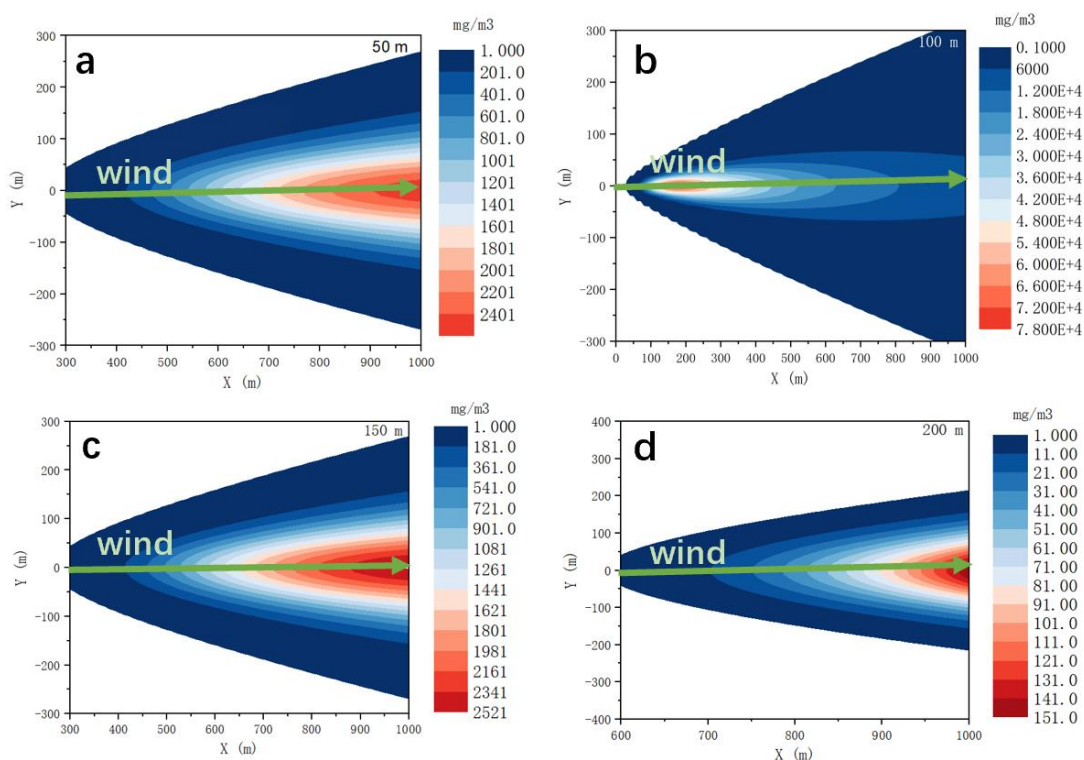


Fig.R2 simulated diffusions of CH<sub>4</sub> at different heights, a.50 m; b.100 m; c.150 m and d. 200 m.

**L250:** If the Gaussian plume doesn't account for background, has the dataset been adjusted to make that correction?

Sorry for the incorrect expression this sentence in our origin manuscript, what we actually want to say is that other researchers did not consider the background value as an unknown parameter in their emission quantification model. They usually extract the background value through some statistical analysis methods, for examples, Nassar *et al* has used the OCO-2 data to evaluate CO<sub>2</sub> emission from power plant successfully, the principle of quantify model is Gaussian dispersion, he regarded the value of background concentration of CO<sub>2</sub> referenced by four plausible manually-selected background regions(Nassar et al., 2021). In Andersen's study, the background concentration of mehane was regarded as the sampled concentration that not effected by the plume in each Flight. And we have modified this sentence as follows:

“The background concentration of mehane was regarded as the sampled concentration that not effected by the plume in each Flight.”

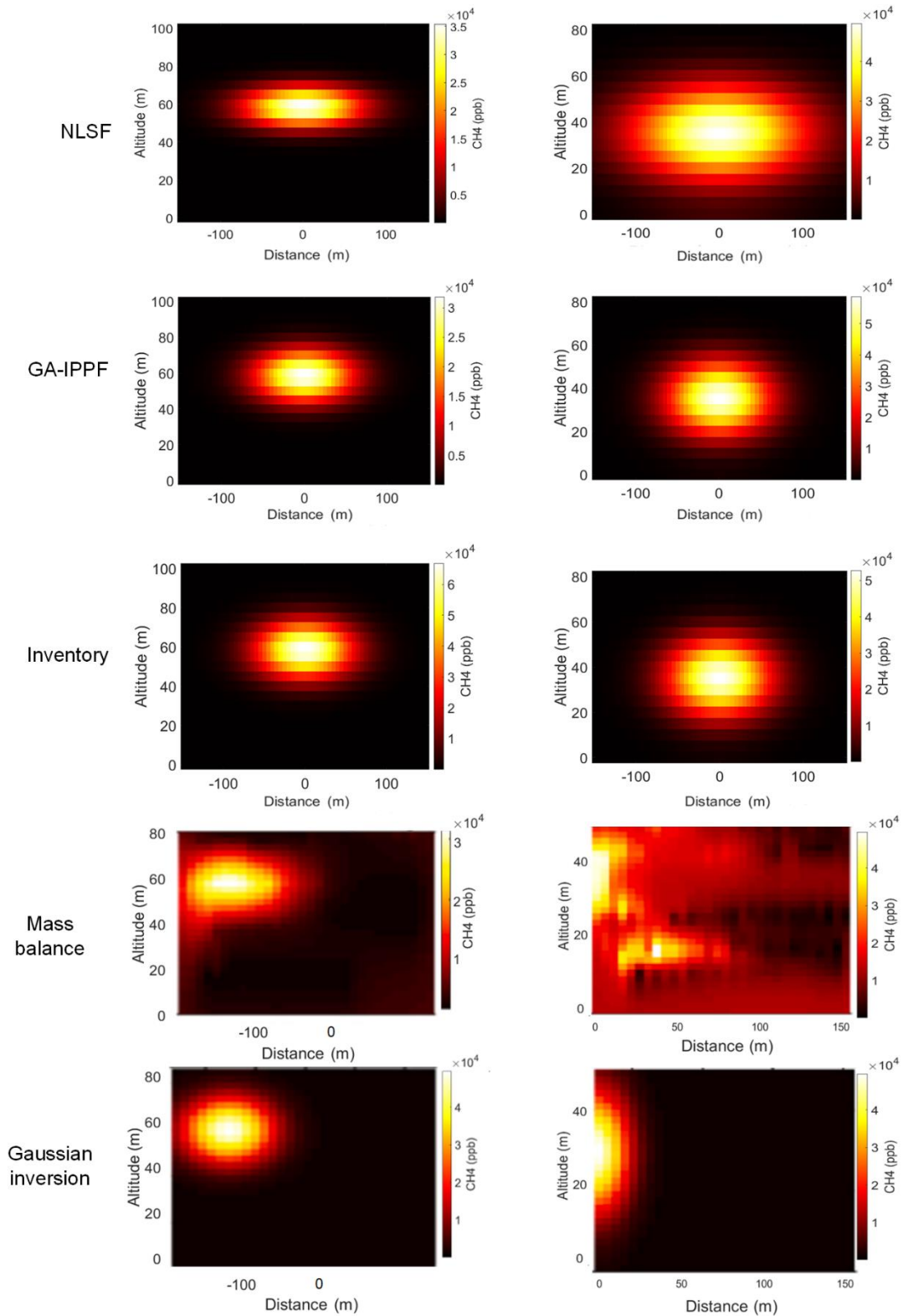
Nassar, R., Mastrogiacomio, J. P., Bateman-Hemphill, W., McCracken, C., MacDonald, C. G., Hill, T., O'Dell, C. W., Kiel, M., and Crisp, D.: Advances in quantifying power plant CO<sub>2</sub> emissions with OCO-2, Remote Sens. Environ., 264, 10.1016/j.rse.2021.112579, 2021.

**Approx L265:** I don't know if this is possible, but it would be incredibly helpful if there was some sort of visualization of the CH<sub>4</sub> atmospheric concentration for each of the methods of quantification (e.g. what does the plume look like in 2-D so that the

variability can be understood). This would be most helpful for where there is significant differences between the methods to show what the plume visualisation looks like in each of the quantifications.

Thank you for this suggestion, we have added this part in our supplement, which is shown as follows:

In this section, we rebuild the 2-D plume of CH<sub>4</sub> dispersion in Flight 6 and Flight 15 based on the different methods, including NLSF, GA-IPPF, inventory, Mass balance and Gaussian inversion. It worth nothing that 2-D plume rebuild by NLSF, GA-IPPF and inventory are according to formula 1. Relative diffusion parameters in inventory 2-D rebuild plume are same as that retrieved by GA-IPPF in each Flight. 2-D plume rebuilt by Masa balance are according to formula 10. 2-D plume rebuilt by Gaussian inversion is according to formula 13.



L330 (approx.) General discussion of measurement stability – the variation in wind parameters, disturbance in the airflow from the drone, changes in plume behaviour are all potential problems for quantification. Are the random errors used here sufficient to capture the potential uncertainty and can they be justified with reference to uncertainty from other studies?

Thanks for this suggestion, variation in wind parameters, disturbance in the airflow from the drone, changes in plume behavior, these items exist in the actual measurements. In the OSSE, firstly, for the variation of wind parameters, we regarded that the random error can represent the uncertainty in the actual emission quantitative evaluation, because we added random errors to the wind speed and wind direction in the simulation experiment, which are randomly distributed within a certain range. For example, if the uncertainty of wind speed is 0.5 m/s, the real value of wind speed is 2.5m/s, wind speed during the measurement may corresponds to 2.2 m/s, 2.8 m/s or 3 m/s. This would represent characteristic of wind speed in the actual measurement. And for the UAV airflow disturbance and plume behavior you mentioned, we thought that they could cause change of the sampling concentration in different locations, thus, we regard the added random error in concentration collection is the combine effect of sample error of Aircore system itself, UAV airflow disturbance and plume change. Therefore, we believe that the random error incorporated in the simulation experiment can be used to assess the uncertainty in the emission quantification.

However, both the solution process in GA and IPPF are not linear, we couldn't define the uncertainty through formula based on uncertainties of the input data. Based on your suggestion, we have also modified the uncertainty calculation method as well as the principle, this suggestion really help us to make our uncertainty evaluation more reasonable, which is shown as follows:

#### “Uncertainty Analyses

The GA-IPPF model will be calculated 1000 times repeatedly based on the collected samples of CH<sub>4</sub> concentration, then, the uncertainty and final retrieved emission rate could be defined by

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (q_i - \bar{q})^2}{N}} \quad (8)$$

$$q_r = \bar{q} = \frac{1}{N} \sum_{i=1}^N q_i \quad (9)$$

$\sigma$  is the uncertainty of retrieved emission rate;  $q_i$  is the  $i$ th retrieved emission rate,  $i=1,2,3 \dots 1000$ ;  $\bar{q}$  is the mean value of the  $q_i$ ;  $N$  is 1000;  $q_r$  is regarded as the value of retrieved emission rate. The values of other parameters ( $a, b, c, d, H, W_s, W_d, B, \alpha$ ) calculated by GA-IPPF are also defined in same principle.”  
L381. The claim that this result would guarantee emission calculation accuracy to better than 99.2% seems very over confident. This type of statement brings me back to the knowledge that methane emissions may be used as legal instruments in the near future and claims of such accuracy when not compared to a blind control is not acceptable. There needs to be a clear rethink of the use of accuracy, error and precision throughout so that it is clear what is being compared. As there is no “known” value, no claim on accuracy can be made.

We are sorry for this simple definition. In origin submission, we declared that the accuracy of GA-IPPF better than 99.2% is based on simulated experiments, which is unreasonable. Therefore, we have selected several release-controlled experiments to evaluate the uncertainty of emission rate calculated by GA-IPPF model, based on the publicly available concentration sampling datasets, as well as meteorological parameters, including wind speed, wind direction and humidity. These release-controlled experiments are similar to the Aircore experiment in Pniówek coal mine, which

are reliable to test the performance of GA-IPPF.

In addition, we also updated the uncertainty analysis method to fully consider the effects of various variables on the final results during the experiments. The specific assessment results are shown as follows:

#### “Uncertainty Analyses

The GA-IPPF model will be calculated 1000 times repeatedly based on the collected samples of CH<sub>4</sub> concentration, then, the uncertainty and final retrieved emission rate could be defined by

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (q_i - \bar{q})^2}{N}} \quad (8)$$

$$q_r = \bar{q} = \frac{1}{N} \sum_{i=1}^N q_i \quad (9)$$

$\sigma$  is the uncertainty of retrieved emission rate;  $q_i$  is the  $i$ th retrieved emission rate,  $i=1,2,3 \dots 1000$ ;  $\bar{q}$  is the mean value of the  $q_i$ ;  $N$  is 1000;  $q_r$  is regarded as the value of retrieved emission rate. The values of other parameters ( $a, b, c, d, H, W_s, W_d, B, \alpha$ ) calculated by GA-IPPF are also defined in same principle.” Thus, we found that the bias of quantified emission rates calculated by GA-IPPF in release-controlled experiment is larger than that in the simulated experiment. In order to express GA-IPPF’s performance rigorously in actual experiment, we presented the uncertainty of quantification results of coal mine Pniówek coal mine shaft in latest submission.

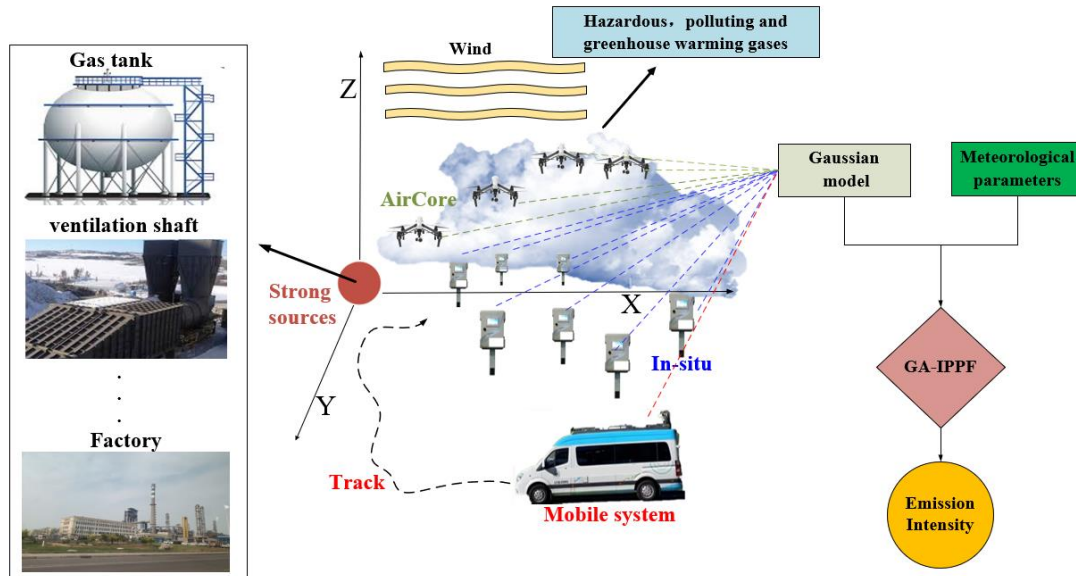
Based on the new result in uncertainty analysis, we also modified this sentence in latest manuscript as follows:

“The UAV-based AirCore system can acquire more than 99 CH<sub>4</sub> samples in actual feasible measurements, therefore, it is believed that accuracy of CH<sub>4</sub> samples (>95.0 %) collected by the AirCore system bring less influence in theory.”

L401. The conclusions feel very generic and non-specific in large parts to the work shown here, they should be reconsidered with the value added of GA-IPPF in mind, rather than generalities of coal emissions. It is good to see that the model is being considered for other uses too, is there any possibility that this could be expanded on in the main text to demonstrate how this would be done for vehicles?

Thank you for your comments on the section of Conclusion, as this manuscript is submitted to special issue of Comet, in original version, we highlighted GA-IPPF and the UAV Aircore system for quantitative assessment of methane emission from coal mining.

In our latest submission version, we presented the framework of GA-IPPF in quantifying gases emission sources, such as factories, gas tank, etc.



**Fig.9.** Application of GA-IPPF in quantifying emission source of gases through different sample systems ; including UAV-based Aircore system, ground-based In-situ network and mobile collection system.

This figure shows target emission sources detectable by GA-IPPF through concentration sampling system based on different platforms, which helps readers to understand the potential usefulness of GA-IPPF.

We also demonstrated that GA-IPPF can perform a quantitative assessment of emissions based on sampling data from UAV-based Aircore system, vehicle-mounted, and ground-based in-situ networks through publicly controlled release experimental datasets. See Table 4.

**Table 4 Performance of GA-IPPF model in different control release experiments**

Database	Number	Gas	Release rates (g/s)	Retrieved by GA-IPPF (g/s)
Agrar Hauser	312_01	CH <sub>4</sub>	0.31±0.03	0.3±0.03
EPA	STR_6061411_01	CH <sub>4</sub>	0.60	0.57±0.04
Prairie Grass	57	SO <sub>2</sub>	101.5	104.7±3.7

Table 4 shows the emission rates and uncertainties through GA-IPPF in control release experiments, and the reported emission rates. The average difference between retrieved emission rates and reported ones is 3.8 %, which indicates the high accuracy of GA-IPPF in quantification estimation.

Therefore, in the latest submission, we have revised the framework of the conclusions and stated the performance of GA-IPPF in different sampling systems as you proposed. The potential application of GA-IPPF is also highlighted in latest Conclusion. Details are shown as follows.

“In this study, we present a quantified model for strong point emission source based on concentration sampling data, named GA-IPPF. During CoMet campaign in 2017, we successfully monitor methane emissions from a ventilation shaft in Pniówek coal mine through the concentration data measured by UAV-based AirCore system. Results show that CH<sub>4</sub> emissions rate from ventilation shaft are not consistent even in a short time.

GA-IPPF can reconstruct the concentration dispersion around the point emission source, and the largest R<sup>2</sup> between the measured CH<sub>4</sub> concentration and the reconstructed concentration in the selected 8 Flights around Pniówek coal mine can reach 0.99.

In Observing System Simulation Experiments (OSSE), we discuss the sensitivity analysis of different parameters setting on final retrieved emission rate by GA-IPPF. We demonstrate that GA-IPPF has self-adjust function to achieve an optimal solution on emission rate, which will reduce the requirements for hardware performance in actual emission quantification experiment.

We also test the performance of GA-IPPF in three control release experiments with different sampling devices, including vehicle-mounted in-situ system, UAV-based AirCore system and ground-based in-situ network observation, and the biases between retrieved emission rates and reported ones within 5.0%.

In future, GA-IPPF has great potential in the point-source quantitation based on mobile concentration sampling system, which can help to renew and enrich the gases emission inventories on strong point sources.”