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Uncertainty Evaluation of Automatic Monitoring System for Fine Particulate Matter in Ambient Air

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Abstract: Ambient air fine particulate matter (PM2.5) is a kind of instantaneous variation of space and time that cannot be repeated. In the process of fluid sampling analysis of real-time data, there are too many variables and too fast component changes for the laboratory to undertake such tests. The fast automatic monitoring technology provides real-time measurement consistency and reliability, which is comparable in terms of cost effectiveness technical process optimization and life cycle. In this case, suitable reference gas for PM2.5 (mg/m^3) could not be found, so β ray or oscillating balance method (X method) and manual weighing method (Y method, as the primary test method) were respectively used. Moreover, related to the sampling frequency of tolerance limit, discussion is given on the detection power (1-β) for the difference (Δ) between the X and Y, for their acceptance probabilistic risk characteristics of operational curve based on the trade-off, and on sample sizes (n) under α and β risks, as well as the acceptable level of the cost of wrong decision. This paper belongs to the research category of unstable samples analysis, and involves the evaluation of two components, $u_{r,\ rel(range)}$ and $u_{R,\ rel(bat)}$. The assessment is based on the overall concept of top-down. All cumulative effects are incorporated into the continuous and closed system as far as possible. Under the premise of ensuring that the acceptable level is under statistical control, reasonable estimates of quality objectives and uncertainties are obtained.

Keywords: Automatic Monitoring System, PM2.5, Site Precision, In-statistical-control, Two Types of Risk, 1-β, Anderson Darling (AD), Top-down Uncertainties

1. Introduction

The majority of people living in China's cities are exposed to unhealthy levels of air pollution, when measured as annual levels of particulate matter, every day. The haze episodes occurred frequently requires setting up and implementing appropriate air quality monitoring systems, in short, quality assurance (QA) and quality control (QC) procedures. The ability to properly plan an air quality monitoring network, implement QA/QC process, as well as disseminate monitoring data and analytical results to stakeholders, should has been developed. About the QA and QC procedures, this paper gives the concrete description as follows.

The sample set/level combination was, routinely accumulated in chronological order over the normal online system operation (X method) and manual weighing method (Y method), estimated for their site precision respectively, without a special treatment for obtaining a better artificially result, so as to leading to the performance underestimated.

Pairs of data of the combination were typically be input using X method and Y method simultaneously in accordance with the procedures outlined in Top-Down Uncertainty Evaluation [1-3], other than bottom-up [4-5], and residuals functions were in turn derived that relates the X method output to Y method. The AD techniques [6] was monitored to continuously demonstrate the proficiency of their residuals that are used for establishing and assuring the data quality, for indicating areas of potential system improvement, analysis rationality of relative variation on data, as well as for being favorable to evaluate the following top-down uncertainties of system in a state of statistical control [7, 8].

Followed by the discussion on sampling frequency of tolerance limit, and estimation of limits of quantification of relative variation [9].

Subsequently, during normal operation of the automatic monitoring system, uncertainty evaluation and recommendations of two components, $u_{r,rel(range)}$ and $u_{R',rel(bat)}$ were given, quality assurance testing is conducted to demonstrate that the agreement between X method and Y method results was kept reasonably and consistently [10-11].

2. Analysis of Experimental Data

PM2.5 level monitoring, for example, in the region of a monitoring station (including sub-stations) in China. The layout of the highest PM2.5 level points selected, use X method and Y method every 12 days, conduct independent sampling and analysis at the same time, and report a representative data pair (24-hour average daily results) [12].

The Table 1 shows 60 representative data pairs collected for three consecutive years in the area where the monitoring station is located, which are used to judge and track the overall level and trend of PM2.5.

Table 1. *PM2.5 data of X method and Y method are statistically summarized.*

The first year										
No	1	2	3	4	5	6	7	8	9	10
month	1		2		3		4		5	
X	72.7	74.3	73.2	73.2	81.4	81.8	61.6	61.5	53.2	53.5
Y	74.8	73.4	75.6	71.4	84.5	83.2	57.5	60.3	52.8	55.1
R (%)	2.8	1.2	3.2	2.5	3.7	1.7	6.9	2	0.8	2.9
bias (%)	-2.8	1.2	-3.2	2.5	-3.7	-1.7	7.1	2	0.8	-2.9
residual	0.9	-2.1	1.2	-3	1.91	0.21	-5.32	-2.42	-1.63	0.37
No	1	2	3	4	5	6	7	8	9	10
month	1		2		3		4		5	
X	81.2	79.1	74.7	81.6	77.9	75.7	61.7	65.1	54.5	47.5
Y	83.1	80.6	72.6	84.7	81.2	80.8	64.4	64.2	57.7	48.3
R (%)	2.3	1.9	2.9	3.7	4.1	6.5	4.3	1.4	5.7	1.7
bias (%)	-2.3	-1.9	2.9	-3.7	-4.1	-6.3	-4.2	1.4	-5.5	-1.7
residual	0.71	0.3	-3.3	1.91	2.1	3.9	1.48	-2.12	1.97	-0.44
No	1	2	3	4	5	6	7	8	9	10
month	1		2		3		4		5	
X	83.8	78.5	78.4	82.2	79.7	85.3	76.4	70.9	66.9	52.1
Y	85.7	82.8	79.3	84.6	80.1	81.9	78.7	76.4	65.3	51.8
R (%)	2.2	5.3	1.1	2.9	0.5	4.1	3	7.5	2.4	0.6
bias (%)	-2.2	-5.2	-1.1	-2.8	-0.5	4.2	-2.9	-7.2	2.5	0.6
residual	0.71	3.1	-0.3	1.21	-0.79	-4.59	1.1	4.29	-2.81	-1.54

Table 1. *Continued.*

The first year										
No	11	12	13	14	15	16	17	18	19	20
month	6	7	8	9	10		11		12	
X	29.9	27.4	23.6	21.8	50.8	46.3	72.4	71.3	80.5	81.4
Y	33	30.9	24.7	24.3	48.7	49.7	70.7	73.9	78.4	80.3
R (%)	9.9	12	4.6	10.8	4.2	7.1	2.4	3.6	2.6	1.4
bias (%)	-9.4	-11.3	-4.5	-10.3	4.3	-6.8	2.4	-3.5	2.7	1.4
residual	1.83	2.23	-0.18	1.22	-3.34	2.16	-2.91	1.39	-3.29	-2.29
No	11	12	13	14	15	16	17	18	19	20
month	6	7	8	9	10		11		12	
X	44.6	36.8	18.4	19.7	58.9	61.6	80.3	82.2	81.7	80.6
Y	43.1	39.9	20.8	20.6	60.1	63.5	79.4	83.3	83.4	82.2
R (%)	3.4	8.1	12.2	4.5	2	3	1.1	1.3	2.1	2
bias (%)	3.5	-7.8	-11.5	-4.4	-2	-3	1.1	-1.3	-2	-1.9
residual	-2.75	1.84	1.11	-0.39	-0.03	0.68	-2.09	-0.09	0.51	0.41
No	11	12	13	14	15	16	17	18	19	20
month	6	7	8	9	10		11		12	
X	45.1	40.2	31.2	22.2	39.6	46.9	82.2	78.9	77.7	79.3
Y	42.4	43.8	36.4	24.4	38.7	50.5	84.5	82.7	78.8	80.4
R (%)	6.2	8.6	15.4	9.4	2.3	7.4	2.8	4.7	1.4	1.4
bias (%)	6.4	-8.2	-14.3	-9	2.3	-7.1	-2.7	-4.6	-1.4	-1.4
residual	-3.95	2.35	3.93	0.92	-2.15	2.36	1.11	2.6	-0.1	-0.1

The estimates given in the table include follows:

The mean values for X method, Y method, R (%) and bias (%) are, 65.7, 66.8, 5.3 and -2.4.

X=1.001Y-1.315, the coefficient of determination 0.99

strongly explains the linear variation trend of X with Y. Since a series of independent data pairs are normally distributed in a bivariate manner (AD=0.831 for residuals in the table), there is no heteroscedasticity treatment for errors under predictive variables (p=0.84, there is a homogeneity of variance diagnosis with 0 and no difference in slope), so data transformation is not considered.

Figure 1 shows the relative variation scatterplot of precision (left) and bias (right) under PM2.5 level. It can be found that the variation of precision and bias in August of the third year is close to ± 15%, so it is necessary to investigate the data submitted in that year. On the whole, the average results of precision and bias given in the table are 5.3% and -2.4$, reaching the expected quality objective, and the system residuals are under the statistical control of 99% probability (AD=0.831), which meets the requirements of the initial stage of quality control activities in the laboratory.

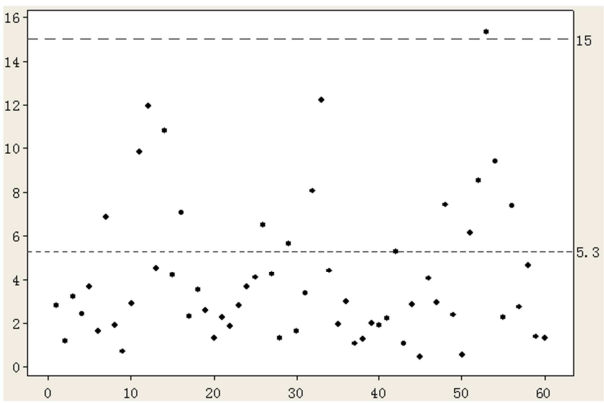

Figure 1. *Scatter plots of relative variation in precision and bias.*

Discussion on sampling frequency of tolerance limit

Figure 2 plots the detection power (1-β) for the difference (Δ) between the X and Y methods, which is an operational curve of acceptance probabilistic risk characteristics based on the trade-off between risk and cost.

Based on different sample sizes (n), this case discusses the sampling frequency under two types of risks (α and β) and the acceptable level of the cost of wrong decision.

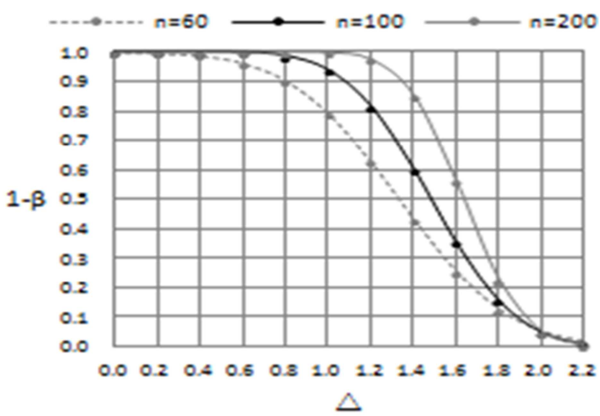

Figure 2. *Corresponding to the risk curve of 1-β under n.*

n in Figure 2 can be expressed as follows:

n=60 (no less than 22 data pairs are sampled each year, equivalent to 1 data pair every 12 days);

n=100 (no less than 44 data pairs are sampled each year, equivalent to 1 data pair every 6 days);

n=200 (no less than 66 data pairs are sampled each year, equivalent to 1 data pair every 3 days).

Although the frequency of use (n=60) in the table saves resources and the Δ values (1.70 to 0.74) obtained at the 90% bilateral interval are within the maximum allowable ± 2 range (equivalent to the intercept 0 test of X=1.001Y-1.315), Figure 2 shows that the failure risk of the 1-β is greater than 10% (the risk of failure is less than 10% for n=100 and n=200).

Therefore, under the condition of the maximum allowable bias Δ value and the cost trade-off, the sampling frequency of once every 6 days is recommended in this paper, which is also helpful to reduce the upper limit of the quality target.

Estimation of limits of quantification of relative variation

Relative variation at too low a level tends to be large. Therefore, in view of the horizontal interval in the table and its relative variation, this paper estimated the limit of quantification of the relationship between the two. This estimation idea is very similar to that of EPA as shown in Figure 3.

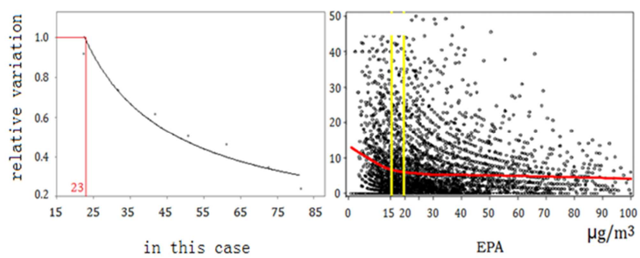

Figure 3. *The case compared with the limit of quantification of EPA data nonlinear regression.*

It can be seen in Figure 3 that the variation at the high level tends to be consistent (≤15%), with the variation at EPA level of 15 (mg/m3) greater than 20 (mg/m3), but the upper limit difference between the two is not significant.

In this case, it is assumed that the limit of quantification is 10% (standard deviation/level=1). According to the ascending level in the table and its corresponding relative variation, the limit of quantification obtained by statistical fitting of the power function is $17.85^{1.08} = 23 \approx \frac{20\mu g}{m^{3}}$. This case suggests that PM2.5 monitoring in the area where the monitoring station is located can be included without considering data below 20 (mg/m3), so as to avoid losing the monitoring of bias due to being too close to the system threshold.

Uncertainty component evaluation

In view of the rationality of the above analysis, the formula given in the table can be used to obtain $\overline{R}_{rel} = 5.3\%$, and $u_{r,rel(range)} = \frac{5.3\%}{1.128} = 4.7\%$.

Bias checks independently confirmed by the measurement system include: internal and external air tightness, zero span, filter membrane, flow rate, temperature and pressure calibration, etc., all of which are directly related to PM2.5 performance monitoring. However, this case focused more on the variation of flow rate adjustment under the correct flow rate of the sampler, namely another component $u_{R',rel(bat)}$.

The identification of PM2.5 comes from the inlet tube and the separator of internal flow rate control system. Since its change directly affects the change of the actual volume flow rate of PM2.5, it is required to be fixed on the two set indexes: one is the variation of the soap film flow rate and 16.67 (L/min) ≤±5%. The other is that the variation between the sampler and the soap film flow rate is ≤±4%. This case uses the soap film flowmeter after the monthly quality inspection to carry out the following flow transfer and calibration on the field sampler:

Unlock the instrument keyboard to enter flow calibration mode, remove the sample top particle cutter and plug into the flowmeter. Record the measured value after the flowmeter reading stabilizes. Enter the average value in the calibration mode for saving, exit and enter the normal measurement mode, remove the flowmeter and install the particulate cutter, and the calibration is over.

Given that the flow adjustment in the table meets the requirements, there is no need to perform further multi-point calibration confirmation. The results of the flow debugging data are subject to the assumption of normal distribution of series residuals (see Figure 4), which are derived from the regression analysis of bivariate random distribution (X=1.015Y-0.357). The analysis conclusions in Figure 4 are as follows:

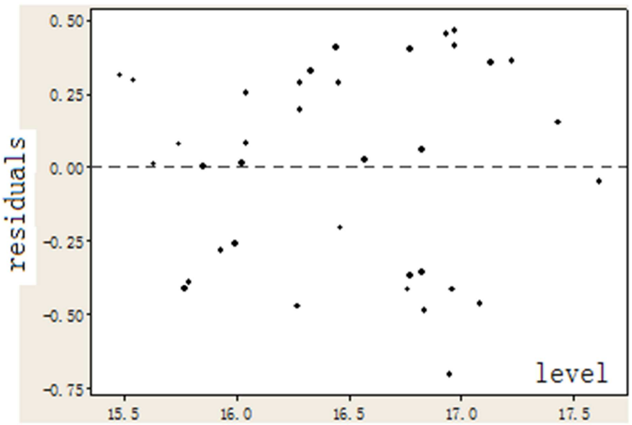

Figure 4. *Residual distribution diagram at the level of Y method.*

The sum of residuals goes to 0 where is no extreme value of residuals. As the level of Y method increases, no curvature shape and longitudinal scatter distribution are found in the residual plot. Accept the null hypothesis of AD statistics with 99% probability.

The above conclusion supports the choice of constant model X=1.015Y-0.357 (but does not consider the adjustment of the sensor of the sampler velocity by regression analysis).

To sum the above and the statistics in Table 2, the monitoring station believes that the uncertainty component is $u_{r,rel(range)} = \frac{5.3\%}{1.128} = 4.7\%$, without regard to the uncertainty contribution of the calibration and $u_{R',rel(bat)}$.

Table 2. *Representative measurements for instantaneous flow calibration (operating conditions).*

The first year												
month	1	2	3	4	5	6	7	8	9	10	11	12
Sampler system	17.07	15.91	16.25	17.55	15.88	16.09	16.63	16.04	15.96	16.06	16.36	15.72
Soap film flowmeter	16.76	16.44	16.77	16.95	16.02	15.93	16.27	16.45	16.28	15.77	16.93	15.85
R (%)	1.8	-3.2	-3.1	3.5	-0.9	1	2.2	-2.5	-2	1.8	-3.4	-0.8
residual	-0.42	0.41	0.4	-0.7	0.02	-0.28	-0.47	0.29	0.2	-0.41	0.45	0
AD (i)	-6.36	-14.9	-23.4	-32.1	-39	-45.2	-53.2	-57.8	-62.7	-67.6	-67.3	-71.55
The second year												
Month	1	2	3	4	5	6	7	8	9	10	11	12
Sampler system	16.65	16.43	17.21	15.66	16.66	15.87	15.11	17.44	16.06	17.17	15.03	15.88
Soap film flowmeter	16.82	16.57	16.83	16.04	17.13	16.28	15.54	17.08	15.79	17.43	15.48	16.33
R (%)	-1	-0.8	2.3	-2.4	-2.7	-2.5	-2.8	2.1	1.7	-1.5	-2.9	-2.8
residual	0.06	0.02	-0.48	0.25	0.36	0.29	0.3	-0.46	-0.39	0.16	0.31	0.33
AD (i)	-69.22	-56.32	-52.48	-49.07	-51.42	-51.63	-48.81	-48.85	-50.46	-47.1	-41.13	-27.29
The third year												
Month	1	2	3	4	5	6	7	8	9	10	11	12
Sampler system	16.75	16.39	17.56	17.07	16.44	15.49	17.03	15.53	16.55	15.83	17.27	16.13
Soap film flowmeter	17.22	16.97	17.61	16.82	16.97	15.63	16.77	15.74	16.46	16.04	16.96	15.99
R (%)	-2.7	-3.4	-0.3	1.5	-3.1	-0.9	1.6	-1.3	0.5	-1.3	1.8	0.9
residual	0.36	0.46	-0.05	-0.36	0.41	0.01	-0.37	0.08	-0.2	0.09	-0.41	-0.26
AD (i)	-23.29	-22.97	-19.82	-19.11	-18.18	-16.48	-16.88	-15.37	-13.99	-13.7	-12.18	-7.85

The statistics of table 2 gives the following:

The mean values for sampler system is 16.35, and soap film flowmeter 16.50.

Relative variation (set indicators) for soap film flowmeter and R (%) is respectively -1.0% (±5%) and -0.7% (±4%).

Uncertainty evaluation and recommendations

By $u_{r,rel(range)} = 4.7\%$, we can directly get $U_{rel} = 2u_{r,rel(range)} = 9.4\%$ and ignore $u_{R',rel(bat)}$.

3. Conclusion

Suggestions for this case are as follows:

1) The three-year accumulation of one data pair every 6 days is conducive to the optimization of quality objectives and the estimation of uncertainty [13].

2) The study of relative variation (≤10%) and limit of quantification is helpful to avoid the influence of excessive variation on quality objective and uncertainty [14].

3) The estimation $U_{rel} = 9.4\%$ is not invariable and needs to be monitored continuously for a long time, which is helpful to make the right quality objective decision [15].

4) Based on the overall concept of top-down, the unified monitoring of the variation trend of various resources is beneficial for remote diagnosis of different sites and multiple devices to maintain an acceptable decision error level [16].

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