State of California Air Resources Board

Method 104

Determination of Beryllium Emissions From Stationary Sources

> Adopted: June 29, 1983 Amended: March 28, 1986

195S/033H

METHOD 1-104 – REFERENCE METHOD FOR DETERMINATION OF BERYLLIUM EMISSIONS FROM STATIONARY SOURCES

1. Principle and Applicability:

1.1 Principle:

Beryllium emissions are isokinetically sampled from the source, and the collected sample is digested in an acid solution and analyzed by atomic absorption spectrophotometry.

1.2 Applicability:

This method is applicable for the determination of beryllium emissions in ducts or stacks at stationary sources. Unless otherwise specified, this method is not intended to apply to gas streams other than those emitted directly to the atmosphere without further processing.

2. Apparatus:

2.1 Sampling Train.

A schematic of the sampling train used by EPA is shown in Figure 104-1. Commercial models of this train are available, although construction details are described in APTD-0581¹, and operating and maintenance procedures are described in APTD-0576. The components essential to this sampling train are the following:

- 2.1.1 Nozzle. Stainless steel or glass with sharp, tapered leading edge.
- 2.1.2 Probe. Sheathed Pyrex² glass. A heating system capable of maintaining a minimum gas temperature in the range of the stack temperature at the probe outlet during sampling may be used to prevent condensation from occurring.
- 2.1.3 Pitot tube. Type S (Figure 104-2), or equivalent, with a coefficient within 5 percent over the working range, attached to probe to monitor stack gas velocity.

¹ These documents are available for a nominal cost from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22151.

² Mention of trade names on specific products does not constitute endorsement by the Air Resources Board.

- 2.1.4 Filter holder. Pyrex glass. the filter holder must provide a positive seal against leakage from outside or around the filter. A heating system capable of maintaining the filter at a minimum temperature in the range of the stack temperature may be used to prevent condensation from occurring.
- 2.1.5 Impingers. Four Greenburg-Smith impingers connected in series with glass ball joint fittings. The first, third and fourth impingers may be modified by replacing the tip with a ½ inch l.d. glass tube extending to one-half inch from the bottom of the flask.
- 2.1.6 Metering Systems. Vacuum gauge, leakless pump, thermometers capable of measuring temperature to within 5°F, dry gas meter with 2 percent accuracy, and related equipment, described in APTD-0581, to maintain an isokinetic sampling rate and to determine sample volume.
- 2.1.7 Barometer. To measure atmospheric pressure to ± 0.1 in Hg.
- 2.2 Measurement of stack conditions (stack pressure, temperature, moisture and velocity).
 - 2.2.1 Pitot tube: Type S, or equivalent, with a coefficient within 5 percent over the working range.
 - 2.2.2 Differential Pressure Gauge. Inclined manometer, or equivalent, to measure velocity head to within 10 percent of the minimum value.
 - 2.2.3 Temperature gauge. Any temperature measuring device to measure stack temperature to within 5°F.
 - 2.2.4 Pressure gauge. Pitot tube and inclined manometer, or equivalent, to measure stack pressure to within 0.1 in Hg.
 - 2.2.5 Moisture determination. Wet and dry bulb therometers, drying tubes, condensers, or equivalent, to determine stack gas moisture content to within 1 percent.
- 2.3 Sample recovery.
 - 2.3.1 Probe cleaning rod. At least as long as probe.
 - 2.3.2 Leakless glass sample bottles. 500 ml.

- 2.3.3 Graduated cylinder. 250 ml.
- 2.3.4 Plastic jar. Approximately 300 ml.

2.4 Analysis

- 2.4.1 Atomic absorption spectrophotometer. To measure absorbance at 234.8 nm. Perkin Elmer Model 303, or equivalent, with N₂O/acetylene burner.
- 2.4.2 Hot plate.
- 2.4.3 Perchloric acid fume hood.

3. Reagents.

- 3.1 Stock Reagents.
 - 3.1.1 Hydrochloric acid concentrated.
 - 3.1.2 Perchloric acid. Concentrated, 70 percent.
 - 3.1.3 Nitric acid. Concentrated.
 - 3.1.4 Sulfuric Aid. Concentrated
 - 3.1.5 Distilled and deionized water.
 - 3.1.6 Beryllium powder. 98 percent minimum purity.

3.2 Sampling.

- 3.2.1 Filter. Millipore AA, or equivalent. It is suggested that a Whatman 41 filter be placed immediately against the back side of the millipore filter as a guard against breaking the Millipore filter. In the analysis of the filter, the Whatman 41 filter should be included with the Millipore filter.
- 3.2.2 Silica gel. Indicating type, 6 to 16 mesh, dried at 350°F for 2 hours.
- 3.2.3 Distilled and deionized water.

- 3.3 Sample recovery
 - 3.3.1 Distilled and deionized water.
 - 3.3.2 Acetone. Reagent grade.
 - 3.3.3 Wash acid. 1.1 V/V hydrochloric acid-water.
- 3.4 Analysis.
 - 3.4.1 Sulfuric acid solution, 12 N. Dilute 333 ml of concentrated sulfuric acid 1 to 1 with distilled water.
 - 3.4.2 25 percent V/V hydrochloric acid-water.
- 3.5 Standard Beryllium Solution
 - 3.5.1 Stock Solution. 1 ug/ml beryllium. Dissolve 10 mg of beryllium in 80 ml of 12 N sulfuric acid solution and dilute to a volume of 1000 ml with distilled water. Dilute a 10 ml aliquot to 100 ml with 25 percent v/v hydrochloric acid, giving a concentration of 1 ug/ml. This dilute stock solution should be prepared fresh daily. Equivalent strength (in beryllium) stock solutions may be prepared from beryllium salts as BeCl₂ and BE(NO₃)₂ (98 percent minimum purity).

4. Procedure

- 4.1 Guidelines for source testing are detailed in the following sections. These guidelines are generally applicable; however, most sample sites differ to some degree and temporary alterations such as stack extensions or expansions often are required to insure the best possible sample site. Further, since beryllium is hazardous, care should be taken to minimize exposure. Finally, since the total quantity of beryillium to be collected is quite small, the test must be carefully conducted to prevent contamination or loss of sample.
- 4.2 Selection of a sampling site and minimum number of traverse points.
 - 4.2.1 Select a suitable sampling site that is as close as practicable to the point of atmospheric emission. If possible stacks smaller than 1 foot in diameter should not be sampled.
 - 4.2.2 The sampling site should be at least 8 stack or duct diameters downstream and 2 diameters upstream from any

flow disturbance such as a bend, expansion or contraction. For a rectangular cross-section, determine an equivalent diameter from the following equation:

$$D_e = 2LW = 0$$
 eq. 104-1

Where:

D_e = equivalent diameter

L = length

W = Width

- 4.2.3 When the above sampling site criteria can be met, the minimum number of traverse points is four (4) for stacks 1 foot in diameter or less, eight (8) for stacks larger than 1 foot but 2 feet in diameter or less, and twelve (12) for stacks larger than 2 feet.
- 4.2.4 Some sampling situations may render the above sampling site criteria impractical. When this is the case choose a convenient sampling location and use Figure 104-3 to determine the minimum number of traverse points. However, use Figure 104-3 only for stacks 1 foot in diameter or larger.
- 4.2.5 To use Figure 104-3, first measure the distance from the chosen sampling location to the nearest upstream and downstream disturbances. Divide this distance by the diameter or equivalent diameter to determine the distance in terms of pipe diameters. Determine the corresponding number of traverse points for each distance from Figure 104-3. Select the higher of the two numbers of traverse points, or a greater value, such that for circular stacks the number is a multiple of four, and for rectangular stacks the number follows the criteria of Section 4.3.2.
- 4.2.6 If a selected sampling point is closer than 1 inch from the stack wall, adjust the location of that point to ensure that the sample is taken at least 1 inch away from the wall.

- 4.3 Cross-sectional layout and location of traverse points.
 - 4.3.1 For circular stacks locate the traverse points on at least two diameters according to Figure 104-4 and Table 104-1. The traverse axes shall divide the stack cross sections into equal parts.
 - 4.3.2 For rectangular stacks divide the cross section into as many equal rectangular areas as traverse points, such that the ratio of the length to the width of the elemental areas is between 1 and 2. Locate the traverse points at the centroid of each equal area according to Figure 104-5.
- 4.4 Measurement of stack conditions.
 - 4.4.1 Set up the apparatus as shown in Figure 104-2. Make sure all connections are tight and leak free. Measure the velocity head and temperature at the traverse points specified by Section 4.2 and 4.3.
 - 4.4.2 Measure the static pressure in the stack.
 - 4.4.3 Determine the stack gas moisture.
 - 4.4.4 Determine the stack gas molecular weight from the measured moisture content and knowledge of the expected gas stream composition. A standard Orsat analyzer has been found valuable at combustion sources. In all cases, sound engineering judgment should be used.
- 4.5 Preparation of a sampling train.
 - 4.5.1 Prior to assembly, clean all glassware (probe, impingers, and connectors) by soaking in wash acid for 2 hours. Place 100 ml of distilled water in each of the first two impingers, leave the third impinger empty, and place approximately 200 g of preweighted silica gel in the fourth impinger. Save a portion of the distilled water as a blank in the sample analysis. Set up the train and the probe as in Figure 104-1.
 - 4.5.2 Leak check the sampling train at the sampling site. The leakage rate should not be in excess of 1 percent of the desired sampling rate. If condensation in the probe or filter is a problem, probe and filter heaters will be required. Adjust the heaters to provide a temperature at or above the stack

temperature. However, membrane filters such as the Millipore AA are limited to about 225°F. If the stack gas is in excess of about 200°F, consideration should be given to an alternative procedure such as moving the filter holder downstream of the first impinger to insure that the filter does not exceed its temperature limit. Place crushed ice around the impingers. Add more ice during the test to keep the temperature of the gases leaving the last impinger at 70°F. or less.

4.6 Beryllium train operation.

- 4.6.1 For each run, record the data required on the example sheet shown in Figure 104-6. Take readings at each sampling point at least every 5 minutes and when significant changes in stack conditions necessitate additional adjustments in flow rate.
- 4.6.2 Sample at a rate of 0.5 to 1.0 ft. min. Samples shall be taken over such a period or periods as are necessary to accurately determine the maximum emissions which would occur in a 24-hour period. In the case of cyclic operations, sufficient tests shall be make so as to allow accurate determination or calculation of the emissions which will occur over the duration of the cycle. A minimum sample time of 2 hours is recommended.
- 4.6.3 To begin sampling, position the nozzle at the first traverse point with the tip pointing directly into the gas stream. Immediately start the pump and adjust the flow to isokinetic conditions. Sample for at least 5 minutes at each, traverse point; sampling time must be the same for each point. Maintain isokinetic sampling throughout the sampling period. Nomographs which aid in the rapid adjustment of the sampling rate without other computations are in APTD-0576 and are available from commercial suppliers. Note that standard monographs are applicable only for Type S pitot tubes and air or a stack gas with an equivalent density.

Contact ARB or the sampling train supplier for instructions when the standard nonograph is not applicable.

4.6.4 Turn off the pump at the conclusion of each run and record the final readings. Immediately remove the probe and

nozzle from the stack and handle in accordance with the sample recovery process described in Section 4.7.

4.7 Sample Recovery.

- 4.7.1 (All glass storage bottles and the graduated cylinder must be precleaned as in 4.5.1.) This operation should be performed in an area free of possible beryllium contamination. When the sampling train is moved, care must be exercised to prevent breakage and contamination.
- 4.7.2 Disconnect the probe from the impinger train. Remove the filter and any loose particulate matter from the filter holder and place in a sample bottle. Place the contents (measured to +1 ml) of the first three impingers into another sample bottle. Rinse the probe and all glassware between it and the back half of the third impinger with water and acetone, and add this to the latter sample bottle. Clean the probe with a brush or a long slender rod and cotton balls. Use acetone while cleaning. Add these to the sample bottle. Retain a sample of the water and acetone as a blank. The total amount of wash water and acetone used should be measured for accurate blank correction. Place the silica gel in the plastic jar. Seal and secure all sample containers for shipment. If an additional test is desired, the glassware can be carefully double rinsed with distilled water and reassembled. However, if the glassware is to be out of use more than 2 days, the initial acid wash procedure must be followed.

4.8 Analysis

- 4.8.1 Apparatus preparation. Clean all glassware according to the procedure of Section 4.5.1. Adjust the instrument settings according to the instrument manual, using an absorption wavelength of 234.8 nm.
- 4.8.2 Sample preparation. The digestion of beryllium samples is accomplished in part in concentrated perchloric acid.
 Caution: The analyst must insure that the sample is heated to light brown fumes after the initial nitric acid addition; otherwise, dangerous perchlorates may result from the subsequent perchloric acid digestion. Perchloric acid also should be used only under a perchloric acid hood.

- 4.8.2.1 Transfer the filter and any loose particulate matter from the sample container to a 150 ml beaker. Add 35 ml concentrated nitric acid. Heat on a hotplate until light brown fumes are evident to destroy all organic matter. Cool to room temperature and add 5 ml concentrated sulfuric acid and 5 ml concentrated perchloric acid. Then proceed with step 4.8.2.4
- 4.8.2.2 Place a portion of the water and acetone sample into a 150 ml beaker and put on a hotplate. Add portions of the remainder as evaporation proceeds and evaporate to dryness. Cool the residue and add 35 ml concentrated nitric acid. Heat on a hotplate until light brown fumes are evident to destroy any organic matter. Cool to room temperature and add 5 ml concentrated sulfuric acid, and 5 ml concentrated perchloric acid. Then proceed with step 4.8.2.4.
- 4.8.2.3 Weigh the spent silica gel and report to the nearest gram.
- 4.8.2.4 Samples from 4.8.2.1 and 4.8.2.2 may be combined here for ease of analysis. Replace on a hotplate and evaporate to dryness in a perchloric acid hood. Cool and dissolve the residue in 10.0 ml of 25 percent V/V hydrochloric acid. Samples are now ready for the atomic absorption unit. The beryllium concentration of the sample must be within the calibration range of the unit. If necessary, further dilution of sample with 25 percent V/V hydrochloric acid must be performed to bring the sample within the calibration range.
- 4.8.3 Beryllium determination. Analyze the samples prepared in 4.8.2 at 234.8 nm using a nitrous oxide/acetylene flame. Aluminum, silicon and other elements can interfere with this method if present in large quantities. Standard methods are available, however, to effectively eliminate these interferences (see Reference 5).

Calibration

- 5.1 Sampling Train.
 - 5.1.1 Use standard methods and equipment as detailed in APTD-0576 to calibrate the rate meter, pitot tube, dry gas meter and probe heater (if used). Recalibrate prior to each test series.

5.2 Analysis

5.2.1 Standardization is made with the procedure as suggested by the manufacturer with standard beryllium solution. Standard solutions will be prepared from the stock solution by dilution with 25 percent V/V hydrochloric acid. The linearity of working range should be established with a series of standard solutions. If collected samples are out of the linear range, the sample should be diluted. Standards should be interspersed with the samples since the calibration can change slightly with time.

6. Calculations

- Average dry gas meter temperature, stack temperature, stack pressure and average orifice pressure drop. See data sheet (Figure 104-6).
- 6.2 Dry gas volume. Correct the sample volume measured by the dry gas meter to stack conditions by using equation 104-2.

$$V_{ms} = V_m \frac{T_s}{T_m} \frac{(P_{bar} + \Delta H/13.6)}{P_s}$$

Where:

V_{ms} = Volume of gas sample through the dry gas meter (stack conditions). ft³

V_m = Volume of gas sample through the dry gas meter (meter conditions). ft³

T_s = Average temperature of stack gas, °R

 T_m = Average dry gas meter temperature, °R

P_{bar} = Barometric pressure at the orifice meter, in Hg.

 ΔH = Average pressure drop across the orifice meter, in H_2O .

13.6 = Specify gravity of mercury.

 P_s = Stack pressure, $P_{bar} + static$ pressure, in Hg.

6.3 Volume of water vapor.

$$V_{w_S} = K_w V_{i_S} \underline{T}_{\underline{S}}$$

$$P_s \qquad eq. 104-3$$

Where:

V_{ws} = Volume of water vapor in the gas sample (stack conditions), ft³.

 $K_w = 0.00267 \frac{\text{in Hg-ft}^3}{\text{ml}^{\circ}\text{R}}$, when these units are used

V_{is} = Total volume of liquid collected in impingers and silica gel (see Figure 104-7), ml.

T_s = Average stack gas temperature, °R

 P_s = Stack pressure, $P_{bar} + static$ pressure, in Hg.

6.4 Total gas volume.

$$V_{total} = V_{m_S} + V_{w_S}$$
 eq. 104-4

Where:

V_{total} = Total volume of gas sample (stack condition), ft³.

 V_{ms} = Volume of gas through dry gas meter (stack conditions), ft^3 .

 V_{ws} = Volume of water vapor in gas sample (stack conditions), ft^3 .

6.5 Stack gas velocity.

Use equation 104-5 to calculate the stack gas velocity.

$$(V_s)_{avg} = K_p C_p (\sqrt{\Delta P})_{avg} \sqrt{\frac{(T_s)_{avg}}{P_s M_s}}$$
 eq. 104 - 5

Where:

 $(V_s)_{avg}$ = Average stack gas velocity, feet per second

 K_p = 85.49 <u>ft</u> <u>(lb-inHg)</u> ½, when these units are sec (lb mole- $^{\circ}$ R-inH₂0) used.

C_p = Pitot tube coefficient, dimensionless.

 $(T_s)_{avg.}$ = Average stack gas temperature, °R.

 $\sqrt{(\Delta P)_{avg}}$ = Average square root of the velocity head of stack gas (in H₂O)^{1/2}(see Figure 104-8).

 P_s = Stack pressure, $P_{bar} + static$ pressure, in Hg.

M_s = Molecular weight of stack gas (wet basis), the summation of the products of the molecular weight of each component multiplied by its volumetric proportion in the mixture, lb/lb. mole.

Figure 104-8 shows a sample recording sheet for velocity traverse data. Use the averages in the last two columns of Figure 104-8 to determine the average stack gas velocity from equation 104-5.

6.6 Beryllium collected.

Calculate the total weight of beryllium collected by using equation 104-6.

$$W_t = V_1C_1 - V_wC_w - V_aC_a$$
 eq. 104-6

Where:

W_t = Total weight of beryllium collected, ug.

V₁ = Total volume of hydrochloric acid from step 4.8.2.4, ml.

 C_1 = Concentration of beryllium found in sample, ug/ml.

V_w = Total volume of water used in sampling (impinger contents plus all wash amounts), ml.

C_w = Blank concentration of beryllium in water, ug/ml.

V_a = Total volume of acetone used in sampling (impinger contents plus all).

C_a = blank concentration of beryllium in acetone, ug/ml.

6.7 Total beryllium emissions.

Calculate the total amount of beryllium emitted from each stack per day by equation 104-7. This equation is applicable for continuous operations. For cyclic operations, use only the time per day each stack is in operation. The total beryllium emissions from a source will be the summation of results from all stacks.

$$R = W_t(V_s)_{avg} A_s/V_{total} \times 86,400 \text{ seconds/day/} 10^6 \text{ ug/g}$$
 eq. 104-7

Where:

R = Rate of emission, g/day.

 W_t = Total weight of beryllium collected, ug.

V_{total} = Total volume of gas sample (stack conditions), ft³.

 $(V_s)_{avg}$ = Average stack gas velocity, feet per second.

 A_s = Stack area, ft^2 .

6.8 Isokinetic variation (comparison of velocity of gas in probe tip to stack velocity).

$$I = 100 V_{total}/A_n\theta(V_s)_{avg} \qquad eq. 104-8$$

Where:

I = Percent of isokinetic sampling.

 V_{total} = Total volume of gas sample (stack conditions) ft³.

 A_n = Probe tip area, ft^2

 θ = Sampling time, sec.

 $(V_s)_{avg}$ = Average stack gas velocity, feet per second.

7. Evaluation of results.

- 7.1 Determination of compliance.
 - 7.1.1 Each performance test shall consist of three repetitions of the applicable test method. For the purpose of determining compliance with an applicable national emission standard, the average of results of all repetitions shall apply.
- 7.2 Acceptable isokinetic results.
 - 7.2.1 The following range sets the limit on acceptable isokinetic sampling results: If 90 percent $\leq l \leq$ 110 percent, the results area acceptable; otherwise, reject the test and repeat.

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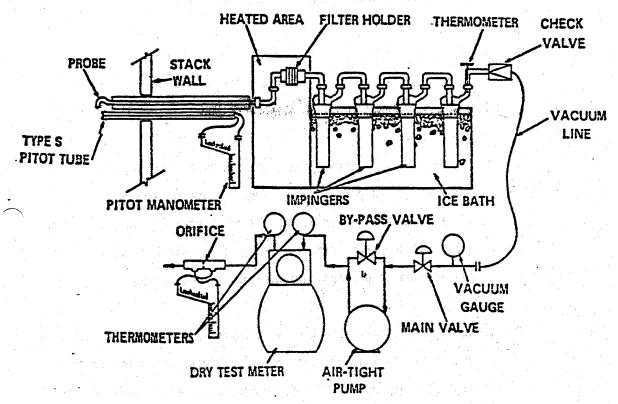
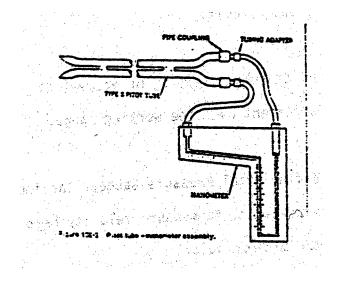


Figure 104-1. Beryllium sampling train



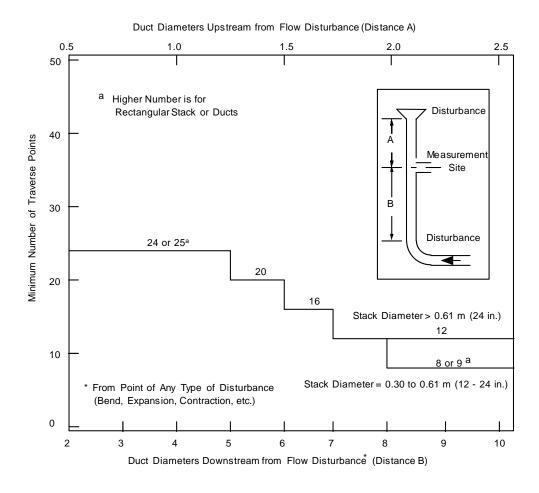


Figure 104-3. Minimum number of traverse points for particulate traverses.

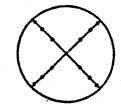


Figure 162-1. Crysta spection of circular stack shawing location of traverse spirits on personal cults diameters.

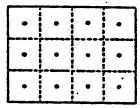


Figure 102-5. Cross section of rectangular stack divided into 12 equal 67343, With Streets seems at contraint of each true.

TABLE 104-1
LOCATION OF TRAVERSE POINTS IN CIRCULAR STACKS
(Percent of stack diameter from inside wall to traverse point)

(Percent of stack diameter from inside wall to traverse point)												
Traverse Point	Number of traverse points on a diameter											
Number on a Diameter	2	4	6	8	10	12	14	16	18	20	22	24
1	14.6	6.7	4.4	3.2	2.6	2.1	1.8	1.6	1.4	1.3	1.1	1.1
2	85.4	25.0	14.6	10.5	8.2	6.7	5.7	4.9	4.4	3.9	3.5	3.2
3		75.0	29.6	19.4	14.6	11.8	9.9	8.5	7.5	6.7	6.0	5.5
4		93.3	70.4	32.3	22.6	17.7	14.6	12.5	10.9	9.7	8.7	7.9
5			85.4	67.7	34.2	25.0	20.1	16.9	14.6	112. 9	11.6	10.5
6			95.6	80.6	65.8	35.6	26.9	22.0	18.8	16.5	14.6	13.2
7				89.5	77.4	64.4	36.6	28.3	23.6	20.4	18.0	16.1
8				96.8	85.4	75.0	63.4	37.5	29.6	25.0	21.8	19.4
9					91.8	82.3	73.1	62.5	38.2	30.6	26.2	23.0
10					97.4	88.2	79.9	71.7	61.8	38.8	31.5	27.2
11						93.3	85.4	78.0	70.4	61.2	39.3	32.3
12						97.9	90.1	83.1	76.4	69.4	60.7	39.8
13							94.3	87.5	81.2	75.0	68.5	60.2
14							98.2	91.5	85.4	79.6	73.8	67.7
15								95.1	89.1	83.5	78.2	72.8
16								98.4	92.5	87.1	82.0	77.0
17									95.6	90.3	85.4	80.6
18									98.6	93.3	88.4	83.9
19										96.1	91.3	86.8
20										98.7	94.0	89.5
21											96.5	92.1
22											98.9	94.5
23												96.8
24												98.9