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DRY DEPOSITION OF SULPHUR DIOXIDE IN THE

ATHABASCA OIL SANDS AREA

by A.H. Jamal, D.S. Chadder, W.A. Murray and R.D. Brymer Promet Environmental Group Ltd. 1338P-36 Avenue" NE CALGARY, Alberta T2E 6T6 Telephone: (403) 276 9123

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ABSTRACT

A field study has been undertaken at the Sandalta lease near Fort HcMurray to determine the sulphur dioxide dry deposition velocities above and within a tree canopy. The concentration gradient method has been used to compute fluxes and dry deposition velocities. The covariance of temperature and vertical wind velocity was measured with a sonic anemometer. The eddy diffusivity for heat was calculated from the ratio of the coveriance to the potential temperature gradient. A flame photometric analyzer was utilized to measure sulphur dioxide concentrations. Most calculations were performed in real time using a digital datalogger. An average daytime dry deposition velocity of 0.8 + 0.7 cm/s was calculated for the September 26, 1983 episode. Sulphur dioxide was observed on nine other occasions during the operational period but because of equipment limitations, it was not possible to calibrate the SO_2 analyzer on the sampling range.

1. INTRODUCTION

Promet Environmental Group **Ltd.** was contracted by Alberta Environment, Research Management Division, to carry out sulphur dioxide flux measurements above and within a tree canopy at Sandalta near Fort HcMurray. The tree canopy consisted of a stand of jack pine, poplar and spruce trees. The project 1s a specific component of an integrated research effort to assess the impact of total acid deposition on terrestrial ecosystems. The experiment required measurements of ambient concentrations of sulphur dioxide and collection of integrated data on the quality and quantity of SO_2 dry deposition. The subsequent field study was also a step toward establishing improved measurement techniques and ensuring stable and consistent operation of all equipment. The raw data were further utilized in the derivation of dry deposition velocities and other flux parameters using the gradient technique. The characteristics and requirements along with comments on the type of results obtained for this and other methods have been presented by Hunt et al (1981). Only the data obtained on September 26, 1983 above the primary tree canopy have been analyzed. The remaining data are presented in raw form in the Appendix.

The field season for the project was to run for four consecutive months commencing 1983 June 01 and ending 1983 September 30. However, due to the developmental nature of the project and the need to devise a sophisticated instrumentation system capable of reliable and accurate data acquisition, the start up of the project was delayed till August. All sensors were operational at this time and a complete set of data was collected on August 26. Thereafter, data for ten days with SO_2 episodes were obtained.

2. METHODS

2.1 SITE SELECTION

The following criteria were necessary for the conditions to be conducive to flux measurements.

- 1. Site should be downwind of an SO_2 source;
- 2. A fetch of uniform surface should extend upwind at least 100 to 200 times the height for which constant flux is assumed;
- 3. The vegetation should be in an actively growing state; and
- 4. Line power should be available.

Figure 1 shows the location of the site for the field study. The SO_2 sources were the Suncor and Syncrude oil sand extraction plants which are licensed for 354 tonnes/day and 292 tonnes/day of sulphur, respectively. The site was situated approximately 25 km to the northeast of the two plants.

An air quality trailer that houses 24 hour continuous ambient monitoring analyzers is located in a 200 square m clearing which is 80 m west of the flux tower. The Monitor Labs analyzers in the trailer measure ambient levels of 0_3 , $S0_2$ and $N0_2$. All four pollutants are measured at two levels, 4 m and 22 m. A weather station situated next to the trailer monitors the following meteorological parameters:

- 1. Precipitation;
- 2. Horizontal wind speed and direction at the 10 and 46 m levels;
- 3. Vertical wind speed at the 46 m level;
- 4. Incoming solar radiation;
- 5. Surface temperature at 1.5 m;
- 6. Surface relative humidity at 1.5 m; and
- 7. Temperature difference between the 10 and 46 m levels.

Figure 1. Topographical map showing project study area.

2.2 EQUIPMENT

There were ten variables recorded continuously during the field experiment at the flux tower. These included:

- 1. Temperature difference over a 3 m height interval at 0.5 m above the primary tree canopy (19 m above ground);
- 2. Temperature difference over a 3 m height interval at 5 m above ground;
- 3. Vertical winds at 8 m and 21 m above ground;
- 4. Ambient SO_2 concentration at heights of 5 m, 10 m, 19 m and 22 m above ground;
- 5. Air temperature at approximately 1 m above ground;
- 6. Relative humidity at about 1 m above ground.

A 24 m tower was used to mount the sensors for items 1 to 4. Copper-constantan thermocouple junctions were used to measure the temperature differences within and above the tree canopy.

A reversing arm mechanism was built to rotate every 15 minutes to help remove any systematic errors associated with the thermocouple sensors. Although based on an original design of Black and MacNaughton (1971), two reversing arm improvements were made by Promet. The first involved the prevention of the booms from over- or under-shooting because of strong winds. This was accomplished by a reversing motor with worm gear drive. Also, special radiation shields were constructed under the direction of T. Gillespie (personal communication), of the University of Guelph, to shield the sensors from radiation influences.

A Campbell Scientific sonic anemometer with a fine wire thermocouple junction $(0.01 \t{mm})$ constructed from chromelconstantan was used for generation of heat flux and eddy correlation data. The fine thermocouple wire measures very minute temperature fluctuations between the ambient air and a reference junction housed inside the anemometer's "thermal mass". The orientation of instrumentation on the tower with respect to the tree canopies is shown in Figure **2.** The reversing arm was mounted facing northwest to reduce the radiative effects of direct solar radiation. The sonic anemometers were directed away from any mechanical air movement caused by the air flow into the sample lines and the reversing of the boom. The sample lines faced southwest, toward the SO₂ sources (i.e., Syncrude and Suncor).

The SO_2 analyser, datalogger and recorders were housed at ground level in a 4 m x 2 m tent. This non-rigid structure was used to minimize the effects of turbulence and radiation which would interfere with the readings of the sensors at the lower level.

A Meloy 8A285E flame photometric detection (FPD) analyzer was used to measure $S0₂$ concentration at four heights. Ambient air was drawn into 6 mm teflon lines continuously by vacuum pumps that provided approximately 1.5 L/m of flow through each of the four sampling lines. All sample lines were of equal length to minimize inconsistencies in the readings due to· adsorption, moisture, and contamination. The sample lines were placed in a 19 mm plastic pipe which was heat traced to prevent condensation in the lines. Four manually operated solenoid zero/span valves were also placed in the pipe. The valves were used to control conditioning of the teflon lines. All four sample lines were conditioned with SO_2 which was delivered under pressure from a cylinder for an average time of 4 minutes for each height, twice a day.

Figure 2 . Schematic showing equipment location on the flux tower. Ambient 502 was sampled at those four levels marked (S), whereas vertical wind speed levels are marked as (SA).

2.3 DATA ACQUISITION

Datalogging and sampling system control were performed using the Campbell Scientific CR7 datalogger. Data were processed in real time by the CR7 and then recorded on magnetic tape. A printer was also used to display the data. All input processing was done over a 15 minute period. That is, every 15 minutes averages and other statistics for each variable were computed and output to the printer and magnetic tape.

Three different scan rates were used to sample the data. Each sensor was scanned at a rate appropriate to its individual response characteristics.

The two sonic anemometers were sampled twice every second. Software in the CR7 computed the eddy correlation of the temperature and vertical winds in real time. The eddy correlation software package in the CR7 contains a subaveraging interval of 180 seconds. Acting like a high band pass filter, eddy frequencies lower than 0.006 Hz are filtered out. SO₂ laden eddies above the forest canopy generally will have frequencies much above this limit (telephone communication with B. Tanner, designer of the sonic anemometer, 1983 June 03).

The CR7 datalogger scanned the output of the SO_2 analyzer once every second. The Meloy SO_2 analyzer attained 95 percent of the SO_2 response in approximately 5 seconds. Only the last five seconds of the 15 second scanning duration were used for computations to ensure that enough time had been allowed for the analyser to respond. Fifteen seconds were spent at each of the four SO_2 sample heights, in succession, before solenoid valves were activated and a new height was sampled. Hence, every minute four 5 second conditional averages were computed and stored in memory. Every 15 minutes, these conditional averages were summed and divided by 15 to provide the average $SO₂$ concentrations for each of the four heights.

seconds: relative humidity, temperature and temperature The following variables were scanned once every ten difference for two levels below and above the tree canopy. Temperature differences were recorded only after the lapse of a five minute sensor equilibrating period following the boom reversal. The boom reversal mechanism and the solenoid valves which select the sample level of ambient air at anyone time were controlled by the CR7 datalogger.

The data accumulated in the CR7 memory were averaged over a 15 minute interval and the output was printed and also recorded on cassette tape. The SO_2 concentrations were also recorded on a Soltec chart recorder. The temperatures of the burner and detector ovens of the SO_2 analyzer were monitored to ensure that the temperatures remained within the recommended operating range. Deviations from this range would result in baseline drift and erratic SO_2 readings.

2.4 CALIBRATION

A two point (zero and span) calibration was performed through each sample line twice a day. Bottled SO_2 gas with a concentration of 79.9 ppbv (210 ug/m^3) traceable to a National Bureau of Standards reference was used.

In addition, multi-point calibrations were performed directly on the analyzers (not through the sampling lines) at least once a day. A Monitor Labs #8550 calibrator with a permeation tube with a permeation rate of 1021 ng/min was used. Five points were used between concentrations of 0 to 1309 ug/ $m³$ (0 to 500 ppbv). To calibrate on the ranges used for sampling (0 to 50 and 0 to 100 ppbv) a permeation tube with a lower permeation rate would have to be used. Unfortunately, a low concentration permeation tube (123 ng/min) was not obtained until September 28, near the end of the study. Summaries of the calibration data are given in the appendix, Section 7.

The two point calibration rather than the multipoints were used in processing the data as there were consistent differences in response between sampling lines, and the two point calibration was on the range (0 to 100 ppbv) actually used for sampling.

2.5 OPERATIONAL PROBLEMS

Once the apparatus was operational, all instruments, with the exception of the sonic anemometer, functioned well for the duration of the project. The sonic anemometer contains two dishes placed 10 cm apart that emit and receive sound waves. Any moisture accumulation on the dishes distorts the signal. This characteristic restricted use of the instrument to the daylight hours as condensation was prevalent at night. The sonic instrument was covered each night to avoid dew or rain on the sensor. The constant handling of the sonic anemometer increased the risk of distorting the alignment of the dishes. A slightly misaligned dish would cause erroneous values for vertical wind speeds. The sonic anemometer mounted at the lower level was not functioning 50 percent of the time due to the above mentioned problems.

The static head exerted by the sampling lines and sample vacuum pump on the intake manifold disturbed the normal operating flow through the SO_2 analyzer. The Meloy analyzer is designed to maintain a constant system flow by drawing air under vacuum across a critical orifice. If the differential pressure across this orifice is changed, the analyzer will show a baseline shift. The CSI-Meloy analyzer system flow is less than 1 L/min. Originally, the sampling pump which was used to draw air into the manifold was set at 25 L/min, however because of the adverse effect it had on the analyzer the sampling flow was trimmed to **1.5** L/min.

The rotameters which were acquired for the higher flow of 25 L/min should be modified to resolve the 1.5 L/min flow better. As well, because of the uncertainty of baseline shifts while sampling ambient air as opposed to $SO₂$ free air, it would be prudent in the future· to provide a multipoint calibration for each of the sampling levels rather than a single SO_2 level.

The equipment in the tent was subject to temperature extremes. During the study period, the temperature in the shelter ranged between -3^oC and 35^oC. The analyzer temperature controller had difficulty keeping up with the ambient temperature variations during this period. The oven and detector temperatures deviated as much as 5 and 10^{0} C from the recommended operating values. The ambient temperature variations also caused changes in the resistances in the voltage divider at the output of the analyzer so that the calibrations varied. To avoid such fluctuations in the operating environment, the possibility of installing a rigid structure with temperature control rather than a tent should be examined. This might possible avoid equipment failures, excessive zero drifts, frequent calibrations and vandalism.

2.6 DATA RECOVERY

Sulphur dioxide was observed on ten days in the August 26 to September 29, 1983 operational period. Unfortunately, except for September 26, the SO_2 analyzer was not calibrated on the range used for sampling because of equipment limitations. The September 26 data were analyzed and are discussed in Section 4, below. Only the raw data are presented in the Appendix for the other episodes.

3. ANALYSIS

3.1 THEORY

Estimates of SO_2 fluxes can be produced using the following equation:

 $F = K_h(So_2 (z_2) - SO_2 (z_1))/(z_2-z_1)$

where,

 $F =$ quantity of SO₂ transferred through unit area per unit time;

 K_h = eddy diffusivity constant (m^2/s) ;

 $SO_2(z)$ = mean SO_2 concentration in ug/m₃ for a specified averaging period at height Z (m);

 z_1 = height of sampling level #1; and

 z_2 = height of sampling level #2.

analogous to that for water vapor or heat fluxes in most The transport mechanism for $S0₂$ is deemed to be atmospheric conditions where turbulent eddies are the dominant means of transport. Heat flux measurements were chosen for the field trials because of the success other researchers had experienced in using fast response sensors such as sonic anemometers.

The following expression shows how $K_h(z)$ was computed:

 K_h = $Z(W^T)^{\prime}$ ($\Delta T + \delta \Delta Z$)

where, WT is the covariance of vertical wind and temperature. That is, the average of the product of the deviations from the means of vertical velocity and temperature. ΔT is the temperature difference over the height interval ΔZ , and δ is the dry adiabatic lapse rate (0.0098 °C/m) .

The deposition velocity is an empirical parameter which is used to estimate SO_2 fluxes from the ambient SO_2 concentrations. Estimates of the deposition velocity (V_d) were computed using the following equation:

$$
V_{A} = F/C
$$

where C is the mean of the SO₂ concentrations at z_2 and z_1 , respectively.

3.2 ERROR CALCULATIONS

The parameters that have been determined in the flux calculations are functions of several variables which are measured independently. It is possible therefore to be able to apply the theory of propagation of error to such cases (Concord 1983:18).

If P is a linear function of j independent variables:

 $P = f (S_1, S_2, S_3, \ldots, S_i)$ Then, the probable error of P is related to the probable errors, Q_1 , $Q_2 \dots Q_j$, of the mean values m_1 , $m_2 \dots m_n$, of the several independent measured quantities S_1, S_2, \ldots, S_j , by the following equation:

$$
Q_p = (\Sigma(dP/ds_j)^2 \cdot Q_j^2)^{1/2}
$$

For example, to calculate the probable error in the deposition velocity, the independent quantities flux and concentration are used:

$$
V_d = F/C
$$

Hence, the probable error is:

$$
Q_V = ((Q_F/C)^2 + (Q_C*F/C^2)^2)^{1/2}
$$

4. RESULTS AND DISCUSSION

On September 26, winds above the tree canopy were light, from the south-southwest, the direction of the Suncor and Syncrude oil sands plants. Skies were clear in the early morning, but it clouded over and was overcast during the SO_2 episode. Sigma theta (standard deviation of azimuthal angle of the wind) values were typical of slightly unstable conditions as shown in Table **1.** Air temperature was 14° C with a relative humidity of 75% at the base of the flux tower.

The average deposition velocity during the episode was $0.8 + 0.7$ cm/s. This is within the range of values determined by other dry deposition experiments over forests. Studies have been undertaken by several other workers to determine the characteristic SO_2 uptake by trees. Most field measurements for $SO₂$ deposition have been made over agriculture land (Chadder et al 1983, Hicks et al 1982, Garland and Bronson 1982)·. Various controlled laboratory measurements of SO_2 fluxes onto forest predict the deposition velocities to be between 0.1 and 0.6 em/sec. Field studies using the eddy correlation technique (Fowler and Cape 1983) determined the rates of dry deposition onto a Scots pine forest to range from 0.05 to 1 cm/s. Similar work by Johansson et al (1982) gave values of V_d of 0.5 cm/s above a birch forest.

The estimated error in the deposition velocity is large, of the order of 0.7 cm/s. This arises from the fact that the velocity is calculated from the ratio of two small differences. Because of the pronounced roughness of the forest surface, the air tends to be well mixed in the vertical and the potential temperature gradients are small. Likewise, the concentration gradients are weak because of vertical mixing of the pollutant.

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In a few cases, upward SO_2 fluxes and negative deposition velocities were calculated. These values may in fact have been positive due to the large error of estimate.

The stand of trees in the current study was a mixture of coniferous and deciduous varieties of varying sizes and shapes. Accordingly, uptake of $S0₂$ will not be as consistent due to the variability of leaf shapes and types found within the canopy.

One of the constraints for the site selection was to have a fetch of uniform surface extending upwind at least 100 times the height of measurement above the tree canopy. This requirement was not met as there existed a 200 m^2 clearing 80 m upwind of the flux tower. This represents a major surface disturbance which will upset turbulent transfer into the canopy.

The calculated SO_2 fluxes underestimate the actual values because eddies with frequencies outside the range 0.006 Hz to 2 Hz are not sampled by the datalogger. McBean (1972) presents a method of correcting flux data for this effect. This would result in a six percent under estimation of flux due to the high frequency cut off and seven to twelve percent under estimation due to the low frequency cut-off. These errors are negligible when compared to the estimated error of up to 100 percent and they therefore have not been taken into account in the calculations.

5. CONCLUSIONS AND RECOMMENDATIONS

The instrumentation performed satisfactorily with the exception of the sonic anemometers. They could only be used during daylight hours since condensation at night distorted the output signal. It is recommended that a more durable vertical wind measuring instrument be used as backup in future studies. so that data can be obtained at night and when it rains.

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The requirement of uniform fetch was not met and may have resulted in apparent upward SO_2 fluxes. Other anomalous results may have been due to horizontal penetration of SO_2 into the canopy.

The average computed dry deposition velocity during the daytime on September 26 was $0.8 + 0.7$ cm/s. Due to the small SO₂ and potential temperature gradients between 19 and 22 m, the probable error in the deposition velocity was relatively high.

Since the study took place during daytime hours, only a small range of meteorological conditions were encountered and no deductions were possible regarding the relationship between deposition velocities and the meteorological conditions.

It is evident that the gradient method for measurements of SO_2 dry deposition velocities and fluxes is limited in accuracy because of the small differences in SO_2 concentration over a tree canopy. Other forest research workers have concentrated on direct eddy measuring techniques rather than gradient methods. However, difficult technical modifications are required to achieve both the high sensitivity and fast response characteristics required in the eddy correlation techniques.

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Table 2. Multipoint Calibration Summaries for August 19, 20,21,26,27,28,29,30 & 31, 1983 والمتواصل والمتواصل والمتعاقب والمتعاقب والمتعارف والمتعاقب والمتعاقب والمتعاقب والمتعاقب والمتعارف ~ 10

H2S carrier gas unless otherwise noted

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Calibrator: Monitor Labs 8500-1769

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Table 4 . $\hspace{1cm}$ Multipoint Calibration Summaries for August 31 & September 1, 8 9, 10, 11 , 12, 14, 15, & 16, 1983

Table 5. Multipoint Calibration Summaries for September 28-29, 1983

Net Responses (mV)												
	S02 Concentration	Date and Time										
	(ug/m3)		Sept. 28/83 Sept. 28/83 Sept. 29/83 1145-1235 MDT 1505-1540 MDT 1305-1338 MDT									
	116	78.5	78.1	82.1								
	103	68.6	67.4	72.0								
	92	61.1	59.0	63.6								
	76	49.9	-47.6	52.4								
	60	39.7	37.0	41.8								
	43	29.6 $\sim 10^{-11}$	26.0	31.3								
	39	27.4	23.6	29.2								
	36	25.9	21.6	27.5								
	32	24.0	20.1	25.3								
	Monitor: Meloy SA285E			Serial $#$: CE003 Range:	0-131 ug/m3							
	Soltec Recorder:		Serial $\#$:	A02615 Range:	$0 - 100$ mV							
	Permeation Rate: 123 ng/min.	Datalogger: Campbell Scientific CR7 Calibrator: Monitor Labs 8500-1769										

Table 6. Multipoint Calibration Summaries for September 28-29, 1983

	22m Height:			19 m		10 ₁₀ m		5. m	
Date	Time	Ze ro	Span	Zero	Span	Ze ro	Span	Ze ro	Span
1983.09.10 1983.09.11 1983.09.11 1983.09.12 1983.09.13 1983.09.14 1983.09.14 1983.09.15	1310-1421 1100-1200 1200-1240 0900-0945 $1009 - 1110$ 0855-1030 $1248 - 1327$ 0815-0834	4.5 3.0 3.2 1.8 1.7 2.6 2.6 2.1	73.5 79.3 78.9 72.2 66.0 57.3 60.3 71.4	8.0 3.3 3.7 2.2 1.9 3.1 2.9 2.1	74.8 79.2 78.4 72.1 65.7 58.3 60.6 71.3	6.2 3.3 3.2 2.2 2.0 3.0 2.8 2.1	76.2 80.1 80.3 72.6 66.5 59.4 61.5 71.9	5.0 3.5 3.4 2.5 3.1 2.9 2.2	77.5 80.8 81.2 67.3 60.6 62.6 72.7
1983.09.15 1983.09.20 1983.09.21 1983.09.22 1983.09.23 1983.09.24 1983.09.25 1983.09.26 1983.09.27	1450-1535 0820-0905 0819-0900 0810-0845 0735-0815 0819-0849 0830-0905 0740-0813 0740-0825	4.7 4.4 2.0 2.2 1.5 2.2 2.3 2.2 3.6	72.4 57.4 71.0 70.4 70.5 70.8 70.6 70.2 71.5	4.6 4.7 2.2 2.3 2.3 2.2 2.8 2.3 3.8	72.1 58.1 71.1 70.8 71.1 71.4 71.3 70.8 71.6	5.0 4.7 1.9 2.2 2.3 2.0 2.7 2.5 3.8	72.9 59.0 71.5 71.4 71.6 72.0 71.8 71.4 72.2	4.3 5.8 2.2 2.6 2.7 .2.5 3. I 3.2 4.0	73.0 59.8 72.3 72.4 72.9 73.1 73.2 72.8 73.7

Table $7\cdot$ Two point, zero and span (210 ug/m3) calibrations, responses in mV or 100 ppbv analyzers scale.

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 $\sim 10^7$

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Table 9. S02 Flux Data Summary for August 27, 1983

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Table 10. S02 Flux Data Summary for August 28, 1983

Table ll. S02 Flux Data Summary for August 30, 1983

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S02 Flux Data Summary (continued) August 30, 1983

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 $\mathcal{L}^{\text{max}}_{\text{max}}$

Table 13. S02 Flux Data Summary for September 10, 1983

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Table 14. S02 Flux Data Summary for September 15, 1983

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Table 15. SO2 Flux Data Summary for September 16, 1983

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 $\mathcal{A}(\mathcal{A})$ and $\mathcal{A}(\mathcal{A})$ and $\mathcal{A}(\mathcal{A})$

on 50 ppbv scale, 100 ppbv after 1415 MST

 $\sim 10^7$

Table 16. S02 Flux Data Summary for September 22, 1983

Table 17. S02 Flux Data Summary for September 24, 1983

on 50 ppbv scale

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Table 18. S02 Flux Data Summary for September 26, 1983

on 50 ppbv scale until 1215 MST, then, 100 ppbv till 1445.

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