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HYDROGEN GENERATION RATES FOR
TANK FARM APPLICATIONS

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INTRODUCTION

Hydrogen is produced in the tank farm waste by radiolysis of water. To prevent a deflagration accident, the production of H₂ must be limited to ensure that the lower flammability limit (LFL) is not reached in any vapor space in the tank farm. Several controls, which protect safe times to reach the lower flammability limit (TLFL) in specific process areas or vessels, depend upon maintaining a defendable maximum H₂ generation rate. This document summarizes the arguments that defend the above values as the maximum rates that will be seen in each of these areas based on the current AB controls, including those in the tank farm Waste Acceptance Criteria (WAC), Chapter 3 (Hazard and Accident Analysis) of the Safety Analysis Report (SAR) and the Technical Safety Requirements (TSR's). Note that there are other organic species that will contribute to the combined Lower Flammability Limit (CLFL), however, this report only addresses the hydrogen generation rate contribution.

DISCUSSION

Hydrogen Generation Rate Calculations

The hydrogen generation rate for a given waste depends on the radiation dose to the waste and the concentration of any hydrogen scavengers that may be present. Free ions of nitrate (NO₃) and nitrite (NO₂) are such scavengers that serve to decrease the overall production of hydrogen.² An older method of calculating the H₂ generation rate considered only the effect of the NO₃ concentration; however a new methodology has been developed that accounts for the contribution of both ions. The term NO_{eff}, equal to the nitrate concentration plus one-half of the nitrite concentration, is the independent variable in the new methodology. The following is a summary of the new methodology used to calculate the volumetric hydrogen generation rate for a given waste.

The hydrogen generation rate, x, is calculated from the radioactive decay heat using:

$$x = (R_{\beta/\gamma} H_{\beta/\gamma} + R_{\alpha} H_{\alpha}) / 10^6 \quad (1)$$

Where R represents the amount of hydrogen generated per 10⁶ Btu from alpha (α) or beta/gamma (β/γ) decay. This R value depends on the concentration of nitrate and nitrite in the waste and is given by³:

$$R_{\alpha} = 134.7 - 82.3 * [NO_{eff}^{-}]^{1/3} - 13.6 * [NO_{eff}^{-}]^{2/3} + 11.8 * [NO_{eff}^{-}] \quad (2)$$

$$R_{\beta/\gamma} = 48.36 - 52.78 * [NO_{eff}^{-}]^{1/3} + 14.1 * [NO_{eff}^{-}]^{2/3} + 0.572 * [NO_{eff}^{-}] \quad (3)$$

These equations are valid only over the range of 0-8 M NO_{eff}, and include a 10% increase to cover the spread of the data. Constant R values are used above 8 M NO_{eff}, however with the hydroxide concentrations in the current waste these high salt concentrations will not be seen.

SUMMARY

The safety analyses used to establish controls in the F and H tank farms impose certain limitations on the hydrogen generation rates ($\text{ft}^3/\text{gallon waste/hr}$) of waste that can be received, transferred, or processed outside of storage tanks in the farms. Table 1 explains the origin of these limits. This document compares these limits with the calculated hydrogen generation rates that might be achieved by actual waste. These rates are shown to be conservative when compared against maximum allowable rates for any current waste and for expected future wastes accepted into the tank farms. Significant changes in the source term or radioactive heat of future wastes will be evaluated against the Authorization Basis (AB).

Currently no waste in any tank farm (supernate, settled sludge, or slurry) exceeds the transfer line limit of $1.2\text{E-}04 \text{ ft}^3/\text{gallon waste/hr}$. Only settled sludge from Tanks 5F, 6F, 12H, 32H, 35H and 39H would exceed the evaporator bottoms and HDB-8 limit of $1.2 \text{ E } -05 \text{ ft}^3 \text{ H}_2/\text{gal/hr}$. However, a 1:1 sludge slurry mixture (necessary for transfer) would only exceed $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{gal/hr}$ in Tanks 6F, 35H and 39H. Therefore, any sludge slurry transfer from these tanks to an evaporator system or to HDB-8 must be evaluated for impact on the hydrogen generation rates in the transfer area.

Table 1 - Hydrogen Generation Rates for Tank Farm Applications

| Process Area | H ₂ Rate ($\text{ft}^3 \text{ H}_2/\text{gallon waste/hr}$) | Explanation |
|---|---|--|
| Transfer lines and transfer facilities excluding HDB-8 | $1.2 \text{ E } -04$ | This value results from using a fresh reactor waste heat generation rate (10.8 BTU/hr/gal) and credits no hydrogen scavengers ¹ . |
| Evaporator Bottoms | $1.2 \text{ E } -05$ | This rate is an imposed limit in Chapter 3 of the SAR. This limit ensures that the time to reach LFL in the evaporator pot is at least 10 days. |
| HDB-8 | $1.2 \text{ E } -05$ | This number is an imposed limit in Chapter 3 of the SAR. This limit ensures that the time to reach LFL in the HDB-8 complex pump tanks is at least 48 hours. |
| Waste Tanks | N/A See Emergency Response Data (ERD) for actual rates. | N/A |

The heat, H , is related to the amount of radioactivity in the waste by:

$$H_{\text{total}} = \sum_{\alpha} Q_{\alpha} A_{\alpha} + \sum_{\beta/\gamma} Q_{\beta/\gamma} A_{\beta/\gamma}, \quad (4)$$

where Q is heat generated per curie for each isotope and A is the total activity of each isotope.

These calculations are performed for the supernate and solids (sludge or salt) phases in each waste tank and published periodically in the Emergency Response Data (ERD).

Effect of Uncertainties in Waste Chemistry on Hydrogen Generation Rate and TLFL

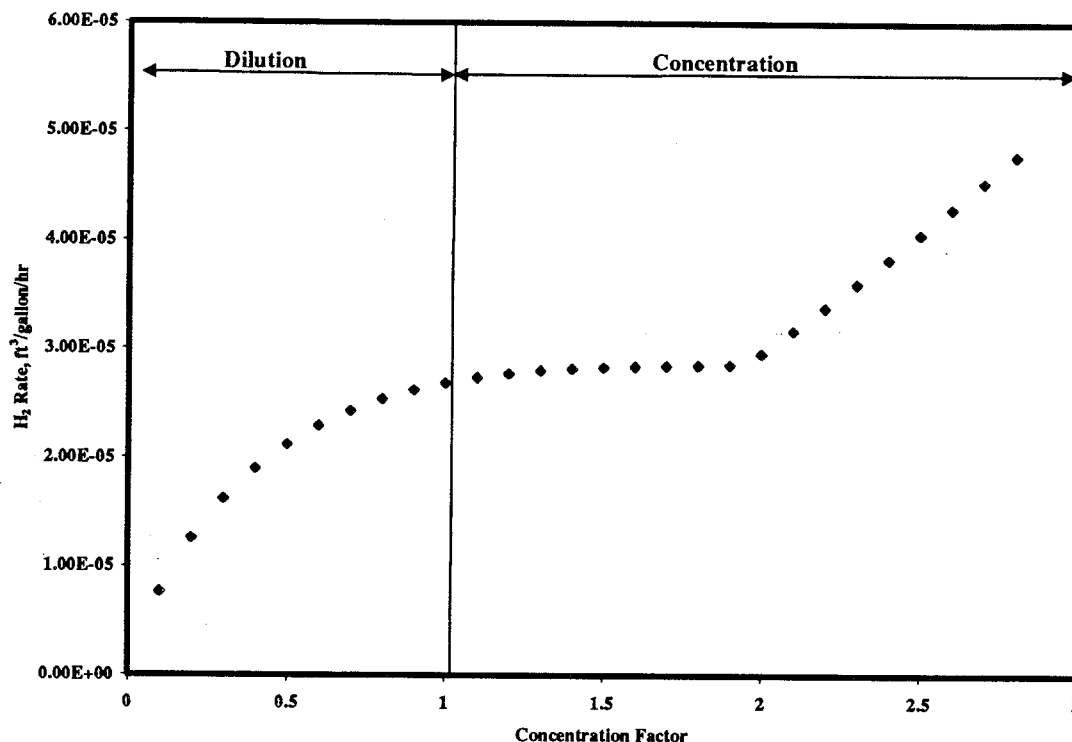
In order to track the proximity of the current waste hydrogen generation rates to the given limits, volumetric hydrogen generation rates will be published in future revisions of the ERD. To account for uncertainties, the volumetric supernate hydrogen generation rates (e.g., those used in waste transfer decisions) in the ERD will include a safety factor of four. This will account for the effect of concentrating waste in the evaporator. The following discussion outlines the conservatism of this safety factor.

Concentration and Dilution of Waste

In the tank farms, there are several processes that result in the concentration and or dilution of the wastes. For example, evaporation concentrates the waste, and transfers often result in waste dilution due to the addition of water or steam. As can be seen from the above equations and discussion, several parameters that are affected by concentration and dilution in the waste contribute to the hydrogen generation rate. The decay heat of the waste, for example, has a direct linear effect on the hydrogen generation rate. As the concentration of nuclides increases, the H_2 rate increases.

The effect of the scavengers on the hydrogen generation rate depends on their solubility, which depends in turn on the hydroxide (OH) concentration. Equations (2) and (3) show the change in the R value as the overall scavenger concentration changes is not linear, but decreases rapidly as the value of NO_{eff} increases. Because the solubility of nitrate and nitrite decreases rapidly as the concentration of OH increases, concentrating waste can have a significant effect on the volumetric hydrogen generation rate, because the scavenger molarity can decrease while the radiation dose per volume increases. Straight dilution only serves to decrease the volumetric hydrogen generation rate. Figure 1 illustrates the varying volumetric hydrogen generation rates for a nominal solution of 3 M OH, 1 M NO_2 and 1 M NO_3 over the range of a 3x concentration to a 0.1x dilution. The heat load for this solution assumes 2 Ci/gal Pu-238, 60 Ci/gal Sr-90 and 60 Ci/gal Cs-137, which is one combination of the nuclides that yields a source term equal to the evaporator bottoms source term limit.

Figure 1 - Effect of Concentration and Dilution on Hydrogen Generation Rate for a 3M OH, 1 M NO₂, 1M NO₃ Solution



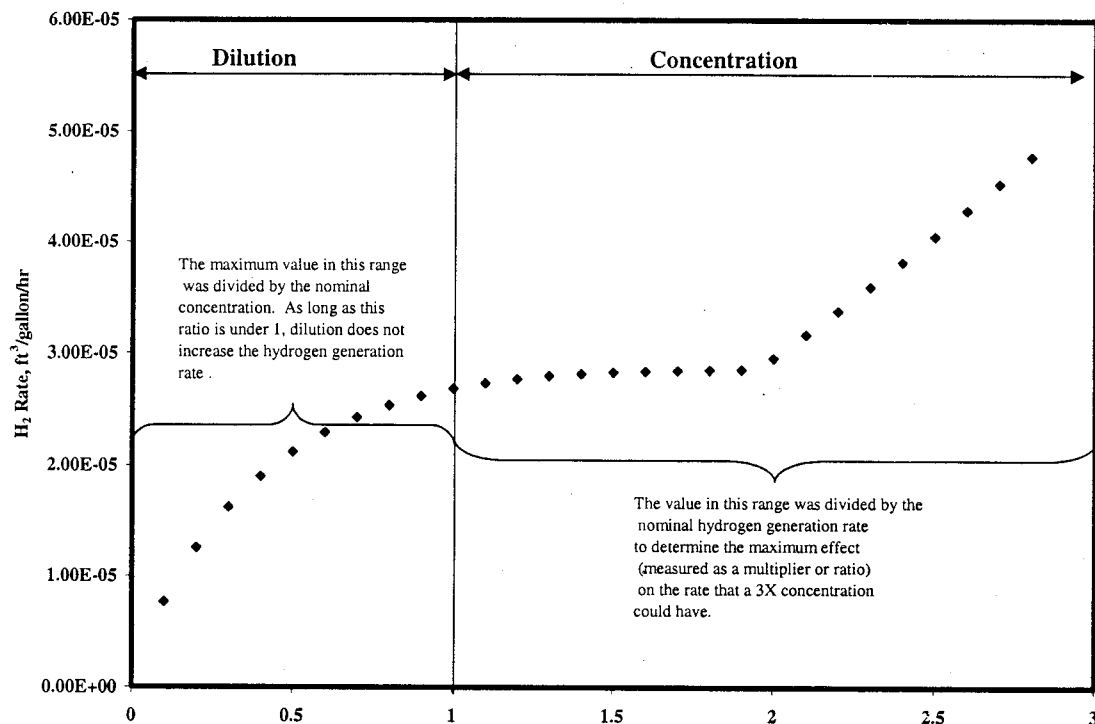
The effect of concentration and dilution of the waste on the volumetric rate is not linear (otherwise Figure 1 would show a straight line). To ensure that multiplying the nominal rate by 4 adequately accounts for the effect, a sensitivity analysis was done to evaluate the impact of concentration and dilution on the volumetric hydrogen generation rate for differing hydroxide and scavenger conditions. The hydrogen generation rates for dilution/concentration factor range of 0.1x to 3x were calculated (as in Figure 1). Although the maximum concentration for some of the feeds into the tank farm are up to 8x, this kind of concentration is not achieved in one pass through the evaporator. A 3x concentration is normal for one pass through an evaporator, and because the feed tank chemistry is updated frequently so that the chemistry reflects any recycles, no higher than 3x needs to be accounted for. At a range of 1 to 8 M OH, both the NO₂ and NO₃ concentrations were varied from 0 to 5 M (these concentrations, except for 0 M, are within the normal operating range of the tank farm) in all possible combinations.

For each combination, the maximum volumetric hydrogen generation rate in the concentration range and the 1x (nominal) concentration was recorded, as well as the maximum volumetric hydrogen generation rate over the dilution range. The ratio of the maximum rates in each range (concentration or dilution) to the nominal rate was determined (See Figure 2 for an explanation of the calculation).

Appendix A contains the results of this analysis and an explanation of the calculation. For each combination of hydroxide and scavenger, the increase in volumetric hydrogen

generation rate due to dilution never exceeded the nominal volumetric rate, confirming that dilution can only decrease volumetric hydrogen production (dilution does increase the overall hydrogen generation rate, but the safety factor accounts for this increase). The hydrogen generation rate in the concentration range averaged 2.5 times the hydrogen rate of the nominal value. For conservatism, the specified volumetric supernate rates calculated in the ERD will be multiplied by 4 to account for the concentration effect in the evaporator. The rates that include this safety factor will be clearly indicated in the ERD. Approximately 10% of the 287 combinations resulted in the hydrogen generation rate at a 3x waste concentration exceeding four times the hydrogen generation rate of the nominal rate. However, none of these exceeded 4.5 times, and it is judged conservative to use a multiplier of 4 given the other conservatism's in the calculation. It should be noted that the measured amounts of hydrogen on tanks 32 and 36 show that our calculations, without the factor of 4 multiplier, are somewhere between 5 and 30 times conservative.⁴

Figure 2 – Explanation of Sensitivity Analysis Calculation



Tank Farm Radionuclide Distribution

Because the hydrogen generation rate is directly related to decay heat, it is important to understand the distribution of the nuclides in the waste as well as which nuclides contribute the most to the hydrogen generation rate. This process knowledge will help defend the maximum hydrogen generation rates for each area.

When fresh acid waste is created in the canyons, NaOH is added to the solution in order to drive the pH above 9.5. This is required by the Tank Farm WAC⁵ to prevent corrosion in the carbon steel tanks. At these high pH values, it is almost impossible for many of the species in the waste to remain in solution. They instead precipitate out, forming the insoluble sludge layer. The supernate is the liquid layer above these settled insoluble species.

The nuclides that remain in solution are mainly Cs-137, a beta emitter, which exists in secular equilibrium with Ba-137_m (1:0.946 ratio). Although the Cs decay to Ba-137_m has negligible energy, the subsequent Ba-137_m decay (which emits a gamma ray) is very intense and gives off significant energy. The supernate hydrogen generation rate is due mainly to this Ba-137_m decay; therefore supernate is considered to release mostly gamma radiation. It has been shown that under alkaline conditions, Pu-238 (an alpha emitter) has limited solubility, so that the majority of the inventory of this species will be found in the sludge.⁶ Sludge is known to have most of the alpha (assumed to be Pu-238) and beta (assumed to be Sr-90 in equilibrium with Y-90 (1:1 ratio)) radiation. Because the decay energy of these three particles (Pu, Sr, and Y) is high, and the nuclides are present in much higher concentration in the sludge, the energy (and therefore the volumetric hydrogen generation rates) of the sludge is generally much higher than those of the supernate. This means that the highest hydrogen generation rates will be seen in pure sludge. Since 100% sludge cannot be pumped, a 1:1 volume ratio of wet sludge to supernate is taken to be the worst case that would be seen in a transfer line (although to actually pump sludge it should be closer to a 1:3 ratio at a maximum value⁷). Pure sludge would also lack water, which is needed as the source of hydrogen in the radiolysis reaction. It is therefore judged that a 1:1 slurry rate is conservative for transfer lines. Using the 1:1 volume ratio adds conservatism in addition to the factor of four in the calculation of the supernate hydrogen generation rates.

TRANSFER LINE AND NON HDB-8 PUMP TANKS MAXIMUM H₂ GENERATION RATE

The hydrogen generation rate $1.2 \text{ E-}04 \text{ ft}^3 \text{ H}_2/\text{gallon/hr}$ originated in a calc note written to evaluate the source term for the New Waste Transfer Facility¹. The calculation used the Fresh Canyon Waste heat value of $10.8 \text{ Btu/gallon/hr}$ at 1 M NO_3 using the NO_3 -only R-Value calculation method⁸ to estimate a maximum hydrogen generation rate for this facility. The heat rate used to derive this number is bounding because it assumes waste from fresh reactor charge. All of the waste in the tank farm today has a lower heat rate than 10.8 Btu/hr/gal . Because the reactors are no longer operating, and the canyon waste is as old as the current waste in the tank farms, it is not expected that any higher heat waste will be received. The receipt of fresh high heat waste would require an evaluation to determine necessary controls to deal with any higher hydrogen generation rates that will be seen. This evaluation would be conducted in accordance with the Waste Acceptance Program and would require an AB change if the hydrogen generation rate exceeded $1.2 \text{ E-}04$ in non HDB-8 complex pump tanks or exceeded $1.2\text{E-}05$ in HDB-8 pump tanks. Table 2 shows the average and maximum volumetric heat rates for both supernate and sludge as of August 1998. These values were calculated from data in WCSysstem, the Tank Farm Waste Characterization Database. The heat rate of 10.8

Btu/hr/gal bounds all of the settled sludge in the tank farm. This heat rate is further conservative, since settled solids must be slurried at least 1:1 with supernate in order to be pumped, and the addition of supernate will reduce the total volumetric heat load.

Table 2 - Current Tank Farm Heat Rates (Btu/hr/gal)

| | Supernate | Sludge | Slurry |
|---------|-----------|--------|--------|
| Average | 0.08 | 1.48 | 0.29 |
| Maximum | 0.34 | 9.34 | 2.61 |

Table 3 lists the volumetric hydrogen generation rates for supernate, settled wet solids, and slurry for each waste tank. These rates are derived from data taken from the Emergency Response Data, calculated as described above. The supernate rates include the 4x safety factor. The Slurry rate is equal to the average of the supernate and the wet solids rate $([\text{supernate rate} + \text{solids rate}]/2)$. Table 3 clearly shows that none of the sludge exceeds the bounding $1.2\text{E-}4 \text{ ft}^3 \text{ H}_2/\text{gallon/hr}$ rate. No sludge slurry (1:1 ratio) exceeds this number, even with the 4x conservatism in the supernate term, so this rate will not be exceeded for any waste in a tank farm transfer line.

Table 3 - Current Tank Farm Volumetric Hydrogen Generation Rates, ft³/gal/hr

| Tank | Supernate* | Sludge Solids | 1:1 Slurry |
|--|------------|---------------|------------|
| 1 | 8.78E-06 | 1.89E-06 | 5.33E-06 |
| 2 | 3.26E-06 | 6.85E-07 | 1.97E-06 |
| 3 | 3.26E-06 | 6.82E-07 | 1.97E-06 |
| 4 | 2.49E-06 | 2.33E-06 | 2.41E-06 |
| 5 | 0.00E+00 | 1.40E-05 | 7.01E-06 |
| 6 | 1.19E-07 | 8.05E-05 | 4.03E-05 |
| 7 | 1.17E-06 | 3.42E-06 | 2.29E-06 |
| 8 | 2.59E-07 | 3.17E-06 | 1.71E-06 |
| 9 | 3.19E-06 | 6.76E-07 | 1.94E-06 |
| 10 | 5.61E-07 | 1.90E-07 | 3.75E-07 |
| 11 | 1.68E-07 | 4.86E-06 | 2.51E-06 |
| 12 | 0.00E+00 | 1.42E-05 | 7.10E-06 |
| 13 | 5.75E-06 | 7.66E-06 | 6.71E-06 |
| 14 | 7.40E-06 | 1.61E-06 | 4.50E-06 |
| 15 | 0.00E+00 | 5.85E-06 | 2.92E-06 |
| 16 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| 17 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| 18 | 1.49E-08 | 3.59E-07 | 1.87E-07 |
| 19 | 8.81E-09 | 8.64E-08 | 4.76E-08 |
| 20 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| 21 | 1.87E-08 | 2.41E-06 | 1.21E-06 |
| 22 | 1.07E-08 | 4.57E-06 | 2.29E-06 |
| 23 | 4.83E-10 | 4.62E-09 | 2.55E-09 |
| 24 | 4.16E-08 | 7.28E-09 | 2.44E-08 |
| 25 | 1.49E-06 | 3.20E-07 | 9.06E-07 |
| 26 | 1.11E-06 | 3.61E-07 | 7.33E-07 |
| 27 | 6.34E-07 | 1.56E-07 | 3.95E-07 |
| 28 | 1.27E-06 | 2.67E-07 | 7.67E-07 |
| 29 | 2.01E-06 | 3.98E-07 | 1.21E-06 |
| 30 | 1.90E-06 | 5.69E-07 | 1.24E-06 |
| 31 | 1.85E-06 | 3.54E-07 | 1.10E-06 |
| 32 | 1.38E-06 | 1.31E-05 | 7.23E-06 |
| 33 | 2.29E-07 | 4.87E-06 | 2.55E-06 |
| 34 | 7.11E-07 | 3.38E-06 | 2.04E-06 |
| 35 | 1.52E-06 | 3.06E-05 | 1.60E-05 |
| 36 | 5.73E-06 | 1.06E-06 | 3.39E-06 |
| 37 | 2.53E-06 | 4.76E-07 | 1.50E-06 |
| 38 | 3.14E-07 | 1.01E-07 | 2.07E-07 |
| 39 | 8.29E-07 | 4.48E-05 | 2.28E-05 |
| 40 | 4.15E-07 | 3.38E-07 | 3.76E-07 |
| 41 | 4.10E-07 | 1.07E-07 | 2.58E-07 |
| 42 | 3.02E-07 | 3.60E-06 | 1.95E-06 |
| 43 | 3.49E-07 | 2.63E-06 | 1.49E-06 |
| 44 | 1.98E-06 | 4.04E-07 | 1.19E-06 |
| 45 | 1.87E-06 | 3.80E-07 | 1.13E-06 |
| 46 | 1.37E-06 | 2.81E-07 | 8.24E-07 |
| 47 | 6.76E-07 | 1.94E-07 | 4.35E-07 |
| 48 | 2.90E-09 | 4.04E-06 | 2.02E-06 |
| 49 | 5.73E-12 | 1.00E-12 | 3.37E-12 |
| 50 | 3.32E-15 | 5.80E-16 | 1.95E-15 |
| 51 | 3.26E-07 | 1.64E-06 | 9.83E-07 |
| * These numbers include a safety factor of four. | | | |

EVAPORATOR MAXIMUM HYDROGEN GENERATION RATES

Chapter 3 of the Safety Analysis Report⁹ for the Tank Farm credits the following control for the Tank Farm Evaporator Explosion Scenario:

Waste transfers into the evaporators shall be controlled such that the bounding evaporator bottoms hydrogen generation rate does not exceed $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{hr}/\text{gal}$. The safety function of this control is to ensure the flammable generation rate in the evaporator remains within the assumed limits.

The value, $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{hr}/\text{gal}$, is the hydrogen generation rate (not considering organic contributions) that would provide greater than 10 days to LFL in the evaporators. The following equation expresses the relationship between TLFL and hydrogen rate:

$$\text{H}_2 \text{ rate} = \frac{0.04 * V_{\text{vapor space}}}{\text{TLFL} * V_{\text{waste}}}$$

The maximum evaporator pot fill limit for the 2F and 2H evaporators is 1950 gallons of material, leaving 2050 gallons of vapor space. Plugging these numbers into the above equations with 456 hours as the time yields the hydrogen generation rate of $1.2\text{E-}5 \text{ ft}^3 \text{ H}_2/\text{hr}/\text{gal}$. Nineteen days is used as the time so that it can be conservatively claimed that ten days in the evaporator will be safe. Ten days is the accepted maximum time required – even in emergency situations – to empty an evaporator. The extra time to reach the LFL was added for conservatism. For the replacement evaporator, with a maximum fill level of 10637 gallons (6540 if equipment is included) and a total volume of 20,000 gallons, the TLFL with this rate is approximately 11 days.

The ten-day limit on the time to CLFL for an evaporator can be maintained by limiting the maximum volumetric hydrogen generation rate of evaporator feed materials. The maximum rate accounts for the concentration effects during evaporation. If, after accounting for all these effects, the maximum rate for a given material is less than $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{hr}/\text{gal}$, then that material can safely be fed to an evaporator.

Referring again to Table 3, it is obvious that:

- Currently, no supernate feed can exceed the limit. The supernate rates are below $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{hr}/\text{gal}$ for all tanks, even with the 4x safety factor.
- Settled solids from Tanks 5F, 6F, 12H, 32H, 35H, or 39H can exceed the limit – if the material could be fed directly to an evaporator, which it cannot. (Of particular interest, settled solids from the existing feed tanks, Tanks 26F and 43H, are below the limit.)
- If the solids were slurried, as they would have to be to make a transfer, only Tanks 6F, 35H or 39H slurry could exceed the limit. These tanks do not serve as feed to the evaporator.

This means that supernate from any tank can be fed to the evaporator without infringing on the safety basis. Any slurry transfer from Tanks 6F, 35H or 39H to an evaporator feed tank would need to be evaluated for its impact on evaporator feed limit.

To aid in the transfer analysis process, the maximum volumetric hydrogen generation rates for each waste tank will be published in the Emergency Response Data. Only transfers to an evaporator feed tank from a tank containing material under the limit will be permitted. Transfers that could potentially exceed the limit will undergo further evaluation. Currently, only sludge slurry from Tanks 6F, 35H or 39H would require evaluation.

HDB-8 AND PUMP TANK MAXIMUM HYDROGEN GENERATION RATE

After re-evaluation of a calculation used to support the HDB-8 safety analysis, it was found that the ventilation system for the pump tanks was not rigorous enough to ensure that the LFL was never reached in the pump tank vapor space.¹⁰ To protect the LFL in the HDB-8 pump tanks, a compensatory measure, allowing a maximum hydrogen generation rate 10 times less than the current bounding rate through HDB-8 ($1.2 \text{ E-}05 \text{ ft}^3/\text{gal/hr}$), was put in to place¹¹. Concentration, Storage and Transfer Engineering judged that this reduction in the hydrogen generation rate would bound any changes resulting from the re-evaluation of this issue. It has since been shown that with this reduced hydrogen generation rate, the pump tanks in HDB-8 have >48 hours to LFL¹². Chapter 3 of the SAR also credits this rate¹³.

The bounding hydrogen generation rate for intended waste transfers into the HDB-8 complex shall be less than or equal to $1.2\text{E-}05 \text{ ft}^3/\text{gal/hr}$. The safety function of this control is to ensure the flammable generation rate in the HDB-8 complex process areas remains within assumed limits.

Using the analysis for the evaporators shows that transfers from Tank 6F, 35H or 39H must be evaluated prior to being sent through HDB-8.

WASTE TANK MAXIMUM HYDROGEN GENERATION RATE

For the waste tanks, there are sludge layers that could have very high volumetric hydrogen generation rates. The transfer line bounding rate applies to slurries, and should not be applied to waste in a tank that could be at a higher concentrations of solids than 50%. Instead, the individual contents of the tank should be considered.

CONCLUSION

The hydrogen generation rates listed in Table 1 are shown to be bounding for the current waste and controls in the Tank Farms, with the exception of Tanks 6F, 35H and 39H slurry which must be evaluated for transfer to an evaporator feed tank or to HDB-8. The evaporator and HDB-8 limit, $1.2\text{E-}05 \text{ ft}^3 \text{ H}_2/\text{gal/hr}$ and the transfer line limit, $1.2\text{E-}04 \text{ ft}^3 \text{ H}_2/\text{gal/hr}$, are well within above the current waste in their applicable process areas. Further, these rates are protected by the WAC and the SAR, respectively. Any

implications of future waste accepted into the tank farms are protected by the Waste Acceptance Program and USQ process.

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Appendix A

This appendix contains all of the results of the sensitivity analysis, the spreadsheet and an explanation of how it was used in the sensitivity analysis, and the Visual Basic Code.

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 1 | 0 | 0 | 3 | 0.3 |
| 1 | 0 | 1 | 1.20 | 0.80 |
| 1 | 0 | 2 | 1.45 | 0.68 |
| 1 | 0 | 3 | 2.14 | 0.47 |
| 1 | 0 | 4 | 2.73 | 0.36 |
| 1 | 0 | 5 | 3.01 | 0.31 |
| 1 | 1 | 0 | 1.56 | 0.60 |
| 1 | 1 | 1 | 1.33 | 0.74 |
| 1 | 1 | 2 | 1.93 | 0.51 |
| 1 | 1 | 3 | 2.58 | 0.38 |
| 1 | 1 | 4 | 3.02 | 0.32 |
| 1 | 1 | 5 | 3.07 | 0.29 |
| 1 | 2 | 0 | 1.45 | 0.66 |
| 1 | 2 | 1 | 1.87 | 0.53 |
| 1 | 2 | 2 | 2.49 | 0.40 |
| 1 | 2 | 3 | 3.03 | 0.32 |
| 1 | 2 | 4 | 3.24 | 0.29 |
| 1 | 2 | 5 | 3.19 | 0.28 |
| 1 | 3 | 0 | 1.93 | 0.51 |
| 1 | 3 | 1 | 2.45 | 0.41 |
| 1 | 3 | 2 | 3.04 | 0.33 |
| 1 | 3 | 3 | 3.40 | 0.28 |
| 1 | 3 | 4 | 3.37 | 0.27 |
| 1 | 3 | 5 | 3.30 | 0.27 |
| 1 | 4 | 0 | 2.47 | 0.40 |
| 1 | 4 | 1 | 3.04 | 0.33 |
| 1 | 4 | 2 | 3.52 | 0.28 |
| 1 | 4 | 3 | 3.54 | 0.25 |
| 1 | 4 | 4 | 3.46 | 0.26 |
| 1 | 4 | 5 | 3.39 | 0.26 |
| 1 | 5 | 0 | 3.04 | 0.33 |
| 1 | 5 | 1 | 3.60 | 0.27 |
| 1 | 5 | 2 | 3.67 | 0.25 |
| 1 | 5 | 3 | 3.59 | 0.25 |
| 1 | 5 | 4 | 3.52 | 0.25 |
| 1 | 5 | 5 | 3.46 | 0.26 |
| 3 | 0 | 0 | 3.00 | 0.30 |
| 3 | 0 | 1 | 1.20 | 0.80 |
| 3 | 0 | 2 | 1.90 | 0.52 |
| 3 | 0 | 3 | 2.80 | 0.36 |
| 3 | 0 | 4 | 3.58 | 0.27 |
| 3 | 0 | 5 | 3.93 | 0.24 |
| 3 | 1 | 0 | 1.56 | 0.60 |
| 3 | 1 | 1 | 1.96 | 0.50 |
| 3 | 1 | 2 | 2.79 | 0.36 |
| 3 | 1 | 3 | 3.66 | 0.27 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 3 | 1 | 4 | 4.19 | 0.23 |
| 3 | 1 | 5 | 4.15 | 0.21 |
| 3 | 2 | 0 | 2.12 | 0.45 |
| 3 | 2 | 1 | 2.80 | 0.35 |
| 3 | 2 | 2 | 3.69 | 0.27 |
| 3 | 2 | 3 | 4.39 | 0.22 |
| 3 | 2 | 4 | 4.33 | 0.20 |
| 3 | 2 | 5 | 4.29 | 0.21 |
| 3 | 3 | 0 | 2.83 | 0.35 |
| 3 | 3 | 1 | 3.67 | 0.27 |
| 3 | 3 | 2 | 4.48 | 0.22 |
| 3 | 3 | 3 | 4.45 | 0.20 |
| 3 | 3 | 4 | 4.40 | 0.20 |
| 3 | 3 | 5 | 4.37 | 0.20 |
| 3 | 4 | 0 | 3.61 | 0.27 |
| 3 | 4 | 1 | 4.48 | 0.22 |
| 3 | 4 | 2 | 4.49 | 0.20 |
| 3 | 4 | 3 | 4.47 | 0.20 |
| 3 | 4 | 4 | 4.45 | 0.20 |
| 3 | 4 | 5 | 4.41 | 0.20 |
| 3 | 5 | 0 | 4.39 | 0.22 |
| 3 | 5 | 1 | 4.47 | 0.20 |
| 3 | 5 | 2 | 4.49 | 0.19 |
| 3 | 5 | 3 | 4.49 | 0.20 |
| 3 | 5 | 4 | 4.47 | 0.20 |
| 3 | 5 | 5 | 4.45 | 0.20 |
| 5 | 0 | 0 | 2.00 | 0.45 |
| 5 | 0 | 1 | 1.17 | 0.82 |
| 5 | 0 | 2 | 1.33 | 0.74 |
| 5 | 0 | 3 | 1.95 | 0.51 |
| 5 | 0 | 4 | 2.50 | 0.39 |
| 5 | 0 | 5 | 2.56 | 0.34 |
| 5 | 1 | 0 | 1.39 | 0.68 |
| 5 | 1 | 1 | 1.37 | 0.71 |
| 5 | 1 | 2 | 1.95 | 0.51 |
| 5 | 1 | 3 | 2.56 | 0.39 |
| 5 | 1 | 4 | 2.63 | 0.33 |
| 5 | 1 | 5 | 2.62 | 0.33 |
| 5 | 2 | 0 | 1.47 | 0.65 |
| 5 | 2 | 1 | 1.96 | 0.50 |
| 5 | 2 | 2 | 2.58 | 0.39 |
| 5 | 2 | 3 | 2.67 | 0.33 |
| 5 | 2 | 4 | 2.66 | 0.33 |
| 5 | 2 | 5 | 2.65 | 0.33 |
| 5 | 3 | 0 | 1.96 | 0.50 |
| 5 | 3 | 1 | 2.56 | 0.39 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 5 | 3 | 2 | 2.67 | 0.32 |
| 5 | 3 | 3 | 2.67 | 0.32 |
| 5 | 3 | 4 | 2.67 | 0.33 |
| 5 | 3 | 5 | 2.66 | 0.33 |
| 5 | 4 | 0 | 2.51 | 0.39 |
| 5 | 4 | 1 | 2.64 | 0.33 |
| 5 | 4 | 2 | 2.66 | 0.33 |
| 5 | 4 | 3 | 2.67 | 0.32 |
| 5 | 4 | 4 | 2.67 | 0.32 |
| 5 | 4 | 5 | 2.67 | 0.33 |
| 5 | 5 | 0 | 2.59 | 0.33 |
| 5 | 5 | 1 | 2.64 | 0.33 |
| 5 | 5 | 2 | 2.66 | 0.33 |
| 5 | 5 | 3 | 2.67 | 0.32 |
| 5 | 5 | 4 | 2.67 | 0.32 |
| 5 | 5 | 5 | 2.67 | 0.32 |
| 8 | 0 | 0 | 1.20 | 0.75 |
| 8 | 0 | 1 | 1.06 | 0.90 |
| 8 | 0 | 2 | 1.01 | 0.98 |
| 8 | 0 | 3 | 1.15 | 0.87 |
| 8 | 0 | 4 | 1.30 | 0.67 |
| 8 | 0 | 5 | 1.30 | 0.67 |
| 8 | 1 | 0 | 1.10 | 0.85 |
| 8 | 1 | 1 | 1.03 | 0.95 |
| 8 | 1 | 2 | 1.15 | 0.87 |
| 8 | 1 | 3 | 1.30 | 0.66 |
| 8 | 1 | 4 | 1.30 | 0.66 |
| 8 | 1 | 5 | 1.30 | 0.66 |
| 8 | 2 | 0 | 1.06 | 0.90 |
| 8 | 2 | 1 | 1.15 | 0.86 |
| 8 | 2 | 2 | 1.30 | 0.66 |
| 8 | 2 | 3 | 1.30 | 0.66 |
| 8 | 2 | 4 | 1.30 | 0.66 |
| 8 | 2 | 5 | 1.30 | 0.66 |
| 8 | 3 | 0 | 1.16 | 0.84 |
| 8 | 3 | 1 | 1.30 | 0.66 |
| 8 | 3 | 2 | 1.30 | 0.66 |
| 8 | 3 | 3 | 1.30 | 0.66 |
| 8 | 3 | 4 | 1.30 | 0.66 |
| 8 | 3 | 5 | 1.30 | 0.66 |
| 8 | 4 | 0 | 1.29 | 0.67 |
| 8 | 4 | 1 | 1.30 | 0.67 |
| 8 | 4 | 2 | 1.30 | 0.66 |
| 8 | 4 | 3 | 1.30 | 0.66 |
| 8 | 4 | 4 | 1.30 | 0.66 |
| 8 | 4 | 5 | 1.30 | 0.66 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 8 | 5 | 0 | 1.29 | 0.67 |
| 8 | 5 | 1 | 1.29 | 0.67 |
| 8 | 5 | 2 | 1.30 | 0.66 |
| 8 | 5 | 3 | 1.30 | 0.66 |
| 8 | 5 | 4 | 1.30 | 0.66 |
| 8 | 5 | 5 | 1.30 | 0.66 |
| 2 | 0 | 0 | 3.00 | 0.30 |
| 2 | 0 | 1 | 1.20 | 0.80 |
| 2 | 0 | 2 | 1.64 | 0.60 |
| 2 | 0 | 3 | 2.41 | 0.41 |
| 2 | 0 | 4 | 3.07 | 0.32 |
| 2 | 0 | 5 | 3.38 | 0.27 |
| 2 | 1 | 0 | 1.56 | 0.60 |
| 2 | 1 | 1 | 1.65 | 0.59 |
| 2 | 1 | 2 | 2.35 | 0.42 |
| 2 | 1 | 3 | 3.09 | 0.32 |
| 2 | 1 | 4 | 3.57 | 0.27 |
| 2 | 1 | 5 | 3.63 | 0.25 |
| 2 | 2 | 0 | 1.82 | 0.53 |
| 2 | 2 | 1 | 2.36 | 0.42 |
| 2 | 2 | 2 | 3.10 | 0.32 |
| 2 | 2 | 3 | 3.73 | 0.26 |
| 2 | 2 | 4 | 3.85 | 0.23 |
| 2 | 2 | 5 | 3.79 | 0.24 |
| 2 | 3 | 0 | 2.42 | 0.40 |
| 2 | 3 | 1 | 3.10 | 0.32 |
| 2 | 3 | 2 | 3.82 | 0.26 |
| 2 | 3 | 3 | 4.04 | 0.23 |
| 2 | 3 | 4 | 3.97 | 0.22 |
| 2 | 3 | 5 | 3.91 | 0.23 |
| 2 | 4 | 0 | 3.09 | 0.32 |
| 2 | 4 | 1 | 3.85 | 0.26 |
| 2 | 4 | 2 | 4.15 | 0.22 |
| 2 | 4 | 3 | 4.09 | 0.22 |
| 2 | 4 | 4 | 4.04 | 0.22 |
| 2 | 4 | 5 | 3.98 | 0.22 |
| 2 | 5 | 0 | 3.82 | 0.26 |
| 2 | 5 | 1 | 4.20 | 0.22 |
| 2 | 5 | 2 | 4.17 | 0.21 |
| 2 | 5 | 3 | 4.13 | 0.21 |
| 2 | 5 | 4 | 4.08 | 0.22 |
| 2 | 5 | 5 | 4.04 | 0.22 |
| 4 | 0 | 0 | 2.50 | 0.36 |
| 4 | 0 | 1 | 1.19 | 0.81 |
| 4 | 0 | 2 | 1.66 | 0.59 |
| 4 | 0 | 3 | 2.44 | 0.41 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 4 | 0 | 4 | 3.12 | 0.31 |
| 4 | 0 | 5 | 3.34 | 0.27 |
| 4 | 1 | 0 | 1.49 | 0.63 |
| 4 | 1 | 1 | 1.72 | 0.57 |
| 4 | 1 | 2 | 2.44 | 0.41 |
| 4 | 1 | 3 | 3.20 | 0.31 |
| 4 | 1 | 4 | 3.48 | 0.26 |
| 4 | 1 | 5 | 3.46 | 0.25 |
| 4 | 2 | 0 | 1.84 | 0.52 |
| 4 | 2 | 1 | 2.45 | 0.40 |
| 4 | 2 | 2 | 3.22 | 0.31 |
| 4 | 2 | 3 | 3.57 | 0.25 |
| 4 | 2 | 4 | 3.55 | 0.25 |
| 4 | 2 | 5 | 3.53 | 0.25 |
| 4 | 3 | 0 | 2.45 | 0.40 |
| 4 | 3 | 1 | 3.20 | 0.31 |
| 4 | 3 | 2 | 3.60 | 0.25 |
| 4 | 3 | 3 | 3.60 | 0.24 |
| 4 | 3 | 4 | 3.58 | 0.24 |
| 4 | 3 | 5 | 3.57 | 0.25 |
| 4 | 4 | 0 | 3.13 | 0.32 |
| 4 | 4 | 1 | 3.57 | 0.25 |
| 4 | 4 | 2 | 3.60 | 0.24 |
| 4 | 4 | 3 | 3.60 | 0.24 |
| 4 | 4 | 4 | 3.60 | 0.24 |
| 4 | 4 | 5 | 3.59 | 0.24 |
| 4 | 5 | 0 | 3.49 | 0.26 |
| 4 | 5 | 1 | 3.56 | 0.24 |
| 4 | 5 | 2 | 3.59 | 0.24 |
| 4 | 5 | 3 | 3.60 | 0.24 |
| 4 | 5 | 4 | 3.60 | 0.24 |
| 4 | 5 | 5 | 3.60 | 0.24 |
| 6 | 0 | 0 | 1.60 | 0.56 |
| 6 | 0 | 1 | 1.14 | 0.84 |
| 6 | 0 | 2 | 1.04 | 0.95 |
| 6 | 0 | 3 | 1.54 | 0.65 |
| 6 | 0 | 4 | 1.92 | 0.50 |
| 6 | 0 | 5 | 1.92 | 0.46 |
| 6 | 1 | 0 | 1.27 | 0.74 |
| 6 | 1 | 1 | 1.08 | 0.90 |
| 6 | 1 | 2 | 1.53 | 0.65 |
| 6 | 1 | 3 | 1.95 | 0.49 |
| 6 | 1 | 4 | 1.95 | 0.45 |
| 6 | 1 | 5 | 1.94 | 0.45 |
| 6 | 2 | 0 | 1.16 | 0.83 |
| 6 | 2 | 1 | 1.54 | 0.64 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 6 | 2 | 2 | 1.96 | 0.49 |
| 6 | 2 | 3 | 1.96 | 0.44 |
| 6 | 2 | 4 | 1.96 | 0.44 |
| 6 | 2 | 5 | 1.95 | 0.44 |
| 6 | 3 | 0 | 1.55 | 0.63 |
| 6 | 3 | 1 | 1.95 | 0.49 |
| 6 | 3 | 2 | 1.96 | 0.44 |
| 6 | 3 | 3 | 1.96 | 0.44 |
| 6 | 3 | 4 | 1.96 | 0.44 |
| 6 | 3 | 5 | 1.96 | 0.44 |
| 6 | 4 | 0 | 1.91 | 0.50 |
| 6 | 4 | 1 | 1.94 | 0.45 |
| 6 | 4 | 2 | 1.95 | 0.44 |
| 6 | 4 | 3 | 1.96 | 0.44 |
| 6 | 4 | 4 | 1.96 | 0.44 |
| 6 | 4 | 5 | 1.96 | 0.44 |
| 6 | 5 | 0 | 1.91 | 0.45 |
| 6 | 5 | 1 | 1.94 | 0.45 |
| 6 | 5 | 2 | 1.95 | 0.44 |
| 6 | 5 | 3 | 1.96 | 0.44 |
| 6 | 5 | 4 | 1.96 | 0.44 |
| 6 | 5 | 5 | 1.96 | 0.44 |
| 7 | 0 | 0 | 1.40 | 0.64 |
| 7 | 0 | 1 | 1.11 | 0.87 |
| 7 | 0 | 2 | 1.02 | 0.97 |
| 7 | 0 | 3 | 1.36 | 0.73 |
| 7 | 0 | 4 | 1.61 | 0.56 |
| 7 | 0 | 5 | 1.61 | 0.54 |
| 7 | 1 | 0 | 1.19 | 0.79 |
| 7 | 1 | 1 | 1.05 | 0.93 |
| 7 | 1 | 2 | 1.36 | 0.73 |
| 7 | 1 | 3 | 1.62 | 0.56 |
| 7 | 1 | 4 | 1.62 | 0.53 |
| 7 | 1 | 5 | 1.62 | 0.53 |
| 7 | 2 | 0 | 1.11 | 0.87 |
| 7 | 2 | 1 | 1.36 | 0.73 |
| 7 | 2 | 2 | 1.62 | 0.56 |
| 7 | 2 | 3 | 1.63 | 0.53 |
| 7 | 2 | 4 | 1.63 | 0.53 |
| 7 | 2 | 5 | 1.62 | 0.53 |
| 7 | 3 | 0 | 1.36 | 0.72 |
| 7 | 3 | 1 | 1.61 | 0.56 |
| 7 | 3 | 2 | 1.62 | 0.53 |
| 7 | 3 | 3 | 1.62 | 0.53 |
| 7 | 3 | 4 | 1.63 | 0.53 |
| 7 | 3 | 5 | 1.63 | 0.53 |

| OH, M | NO ₂ , M | NO ₃ , M | H ₂ @ 3x / H ₂ @ 1x | Max Dilution H ₂ /1x H ₂ |
|-------|---------------------|---------------------|--|---|
| 7 | 4 | 0 | 1.59 | 0.57 |
| 7 | 4 | 1 | 1.61 | 0.54 |
| 7 | 4 | 2 | 1.62 | 0.53 |
| 7 | 4 | 3 | 1.62 | 0.53 |
| 7 | 4 | 4 | 1.62 | 0.53 |
| 7 | 4 | 5 | 1.63 | 0.53 |
| 7 | 5 | 0 | 1.59 | 0.54 |
| 7 | 5 | 1 | 1.61 | 0.54 |
| 7 | 5 | 2 | 1.62 | 0.53 |
| 7 | 5 | 3 | 1.62 | 0.53 |
| 7 | 5 | 4 | 1.62 | 0.53 |
| 7 | 5 | 5 | 1.62 | 0.53 |
| | | | | |
| | | Max Value | 4.49 | 0.98 |
| | | Average Value | 2.47 | 0.45 |

[illegible]

The following are the two Visual Basic Subroutines used in the Sensitivity Analysis:

Function conc(x, Pu, Sr, Cs, No2, No3, OH)

```

heatalpha = Pu * 0.00186 * 60
heatbeta = (0.0000575 + 0.00031) * 60 * Sr _
           + (0.000066 + 0.946 * 0.000224) * Cs * 60
ohnew = x * OH
If ohnew > 10 Then
    conc = "N/A"
    Exit Function
End If
no2new = x * No2
no3new = x * No3
heatanew = x * heatalpha
heatbnew = x * heatbeta
addsol = 8.6 + 0.26 * ohnew - 2.57 * (ohnew) ^ 0.5
If No2 <> 0 And No3 <> 0 Then
    ratio = No3 / No2
    no2sol = addsol / (1 + ratio)
    no3sol = no2sol * ratio
ElseIf No3 = 0 Then
    no2sol = addsol
    no3sol = 0
Else
    no3sol = addsol
    no2sol = 0
End If

If no3sol >= no3new Then
    nitrate = no3new
Else
    nitrate = no3sol
End If
If no2sol >= no2new Then
    nitrite = no2new
Else
    nitrite = no2sol
End If
half = nitrate + 0.5 * nitrite
If half <= 8 Then
    Ralpha = (1.3 - 0.79 * half ^ (1 / 3) - 0.13 * half ^ (2 / 3) + 0.11 * half) * 94.37 * 1.1
    Rbeta = (0.466 - 0.51 * half ^ (1 / 3) + 0.14 * half ^ (2 / 3) + 0.0055 * half) * 94.37 * 1.1
Else
    Ralpha = (1.3 - 0.79 * 8 ^ (1 / 3) - 0.13 * 8 ^ (2 / 3) + 0.11 * 8) * 94.37 * 1.1
    Rbeta = (0.466 - 0.51 * 8 ^ (1 / 3) + 0.14 * 8 ^ (2 / 3) + 0.0055 * 8) * 94.37 * 1.1

```


End If

conc = (heatanew * Ralpha + heatbnew * Rbeta) / 1000000#

End Function

Sub Sensitivity()

For x = 7 To 294

one = Sheets("concentrate").Columns(5).Rows(x)

two = Sheets("concentrate").Columns(6).Rows(x)

three = Sheets("concentrate").Columns(7).Rows(x)

Sheets("Concentrate").Columns(5).Rows(2) = one

Sheets("Concentrate").Columns(6).Rows(2) = two

Sheets("Concentrate").Columns(7).Rows(2) = three

threeex = Sheets("concentrate").Columns(4).Rows(22)

Max = Sheets("concentrate").Columns(4).Rows(23)

dilution = Sheets("concentrate").Columns(4).Rows(20)

concentrate = Sheets("concentrate").Columns(4).Rows(21)

nominal = Sheets("concentrate").Columns(4).Rows(19)

Sheets("Concentrate").Columns(12).Rows(x) = nominal

Sheets("Concentrate").Columns(8).Rows(x) = threeex

Sheets("Concentrate").Columns(9).Rows(x) = Max

Sheets("Concentrate").Columns(10).Rows(x) = dilution

Sheets("Concentrate").Columns(11).Rows(x) = concentrate

Next x

End Sub

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