

Considerations of an Air-Quality Standard to Protect Terrestrial

Vegetation from Acidic Precipitation

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## ABSTRACT

Acidic precipitation is here defined as wet or frozen deposition with a hydrogen ion concentration greater than  $2.5 \mu\text{eq l}^{-1}$ . At the present time there is an inadequate amount of information that shows decreases in crop growth except for one field study. Most studies with plants (crops and forests) are inadequate for standard setting because they are not conducted in the field with adequate randomization of plots coupled with rigorous statistical analyses. Although visible injury to foliage has been documented in a variety of greenhouse studies, no experimental evidence demonstrates loss of field crop value or reduction in plant productivity due to visible foliar injury. Acidic precipitation can contribute nutrients to vegetation and could also influence leaching rates of nutrients from vegetation. Although these processes occur, there are no data that show changes in nutrient levels in foliage that relate to crop or natural ecosystem productivity. Experimental results show that fertilization of ferns is inhibited by current levels of acidic precipitation in the northeastern United States. However, the overall impacts of inhibited fertilization on perpetuation of the species or ecosystem productivity have not been evaluated. Simulated acidic precipitation has been shown to effect plant pathogens in greenhouse and field experiments. Simulated acidic precipitation inhibited pathogen activities under some circumstances and promoted pathogen activities under other circumstances. No conclusion can be drawn about the effects of current levels of precipitation acidity on plant pathogen-host interactions. From these data it must be concluded that research on the effects of acidic precipitation on terrestrial vegetation is too meager to draw any conclusions with regard to an air quality standard. It has yet to be demonstrated that current regional levels

of precipitation acidity alone significantly injure terrestrial vegetation. Although there are some suggestions that crops might be injured, the evidence is not conclusive. More research data need to be evaluated in this area of effects of acidic precipitation on terrestrial vegetation before a standard could be established, if one is necessary.

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Acidic precipitation is here defined as wet or frozen deposition with a hydrogen ion concentration greater than  $2.5 \mu\text{eq l}^{-1}$ . Much of what is known about its effects has been derived from investigations of acid deposition (acidic precipitation and dry deposition) problems in Canada and Scandinavia, as well as the United States. Significant interest has been aroused in both legislative and executive branches of the U. S. Government in determining the severity of problems relating to acidic precipitation. As a result of this interest an air quality standard may be established to limit the concentration or even the deposition of pollutants associated with acidic precipitation to prevent or ameliorate its impacts.

Establishing an air quality standard for precipitation acidity to protect terrestrial vegetation involves several assumptions. The first of these is that acidity in precipitation causes injury to vegetation. Injury can be defined as: a) loss of crop yield and/or quality, b) visible injury which would reduce the market value of a crop, c) loss of forest yield or long-term growth of trees, d) visible injury to ornamental plants that would reduce their aesthetic value, e) substantial alterations of plant community composition leading to ecosystem simplification, f) changes in herbivore populations or communities leading to any of the injuries above, and g) altered sensitivity to other air pollutants and/or plant pathogens leading to any of the injuries above. There is also an assumption that controlling

the acidity of precipitation by establishing a standard which is not allowed to be exceeded will prevent or ameliorate known or highly probable injuries.

The impacts, if any, that acidic precipitation is having on terrestrial plants outside of laboratory or controlled field conditions have yet to be determined. Experimental research to date has shown that plant response can vary greatly depending on the species, the environment, and the method of exposure. Due to this variability, it has been necessary to study the impacts of acidic precipitation on a plant species by plant species basis. From this approach an overall picture of which plant groups are most sensitive and the types of injury which occur is beginning to emerge.

Experimental exposures of plants to simulated acidic precipitation over days, weeks, or a growing season are conducted to evaluate relationships between treatment  $H^+$  concentrations and plant responses. In evaluating experimental results, it is important to distinguish between effects observed at  $H^+$  concentrations which are much above the volume-weighted mean  $H^+$  concentration of ambient precipitation, near ambient  $H^+$  levels, and so-called "control"  $H^+$  levels which are much below ambient levels (e.g.,  $pH \geq 5.6$ ). Both extremes, unusually high and unusually low  $H^+$  concentrations, represent conditions that do not occur in the northeastern United States. However, when a large range of  $H^+$  concentrations are tested, linear or curvilinear functions, with known confidence limits, of plant responses versus  $H^+$  concentration will provide information to estimate changes in plant responses due to ambient or anticipated levels of acidity.

#### EVIDENCE OF INJURY TO TERRESTRIAL VEGETATION

Loss of Crop Yield and/or Food Quality. Exposure to acidity in pre-

precipitation may conceivably decrease yield and/or food quality of crops. Since it is generally accepted that managed soils are less susceptible to perturbations by acidic precipitation due to normal applications of fertilizers and lime,<sup>1</sup> any effects of acidic precipitation on crop plants would probably result from exposure of foliage. Significant reductions (19% and 11%) in dry weight of trifoliolate leaves and dry weights of pods plus seeds, respectively, of bush beans occurred after exposure to acidic mists even though there was no visible leaf injury, when plants were grown in pots in a greenhouse.<sup>2</sup> In contrast, simulated acidic mists of pH 3.0 did not reduce plant growth rates of yellow birch even though all leaves exhibited leaf pitting and curling.<sup>3</sup> With many crop plants yield is determined by the development and survival of reproductive organs as well as by cumulative injury to foliage. Because relationships between growth and yield (plant biomass, economic crop yield, etc.) and visible plant injury, or any other index of injury, remain unknown for most plant species, no unified view of visible injury versus yield impacts is available at present.

Simulated acidic rain of pH 3.1 and below decreased the dry mass of seeds, leaves, and stems of pinto beans grown in pots under greenhouse conditions. On a percentage-mass basis the decrease in seed yield was comparable with reductions in biomass of leaves and stems. This decrease in yield was attributed to both (1) a decrease in the number of pods per plant and, (2) a decrease in the number of seeds per pod.<sup>4</sup> Simulated acidic rain decreased the dry mass of both stems and leaves of soybeans grown in a greenhouse. However, an increase in seed yield occurred when plants were exposed to rain of pH 3.1. A larger dry mass per seed was responsible for the larger dry mass of seeds per plant.<sup>4</sup> Effects of simulated acidic rain on 27 crop plants grown in pots were determined. The marketable yields of five crops (radish, beet, carrot, mustard greens, and broccoli) were reduced, while yields of

six crops (tomato, green pepper, strawberry, alfalfa, orchard grass, and timothy) were increased when they were treated with acidified rain at various pH levels between 3.0 and 4.0 compared to pH 5.7 treatments. No consistent effects were observed for 16 other crops.<sup>5</sup>

Simulated acidic rain caused significant reductions in soybean yield and quality under experimental field conditions using standard agronomic practices.<sup>6</sup> Plants exposed to simulated acidic rainfalls of pH 4.0, 3.1, and 2.7 in addition to ambient rainfalls decreased seed yields 2.6, 6.5, and 11.4%, respectively, compared to plants exposed to ambient conditions only (Fig. 1). On a per-plant basis, total seed protein decreased 10, 19, and 23% in plants exposed to simulated rainfalls of pH 4.0, 3.1, and 2.7, respectively, compared to plants exposed to ambient conditions only (Table 1).<sup>6</sup> At the present time, this is the only adequately replicated experiment known that demonstrates significant alterations in crop productivity and quality under standard agronomic conditions due to precipitation acidity. Additional experimentation of this type using conventional agronomic practices is needed to evaluate injuries that may occur due to precipitation acidity at or close to current acidity levels.

Visible Injury which would Reduce the Market Value of a Crop. If the foliage or fruits of some crops exhibit blemishes, the market value would be reduced substantially. Simulated acidic rain has induced lesions on leaves and reproductive structures. The highest experimental treatment pH at which visible lesions on leaves have been observed is near pH 4.0.<sup>5</sup> A large percentage of the leaf area may exhibit lesions at pH 3.1, but at pH levels above 3.1 less than 5% injury is observed.<sup>2,3,9-14</sup> No relationship between crop yield reduction and foliar injury has been established, so this indicator of injury should not by itself be used to set a standard for precipitation acidity.

There is no evidence that current acidity levels injure field or forest plant foliage. Experiments at low pH, however, indicate that this may be possible. Most injury to foliage by simulated acidic rain occurs on expanding or recently expanded leaves and needles. This is a critical stage in leaf development. Young (14 day old) birch seedlings are much more sensitive than older (6 week old) seedlings.<sup>3</sup> Leaves that had just reached full expansion were more sensitive to simulated acidic rain than unexpanded and already fully expanded birch leaves.<sup>3</sup> Needle elongation was inhibited if simulated acidic rain solutions of pH levels below 3.0 were applied to immature pine fascicles.<sup>9,15</sup> Applications of acidic rain may also inhibit leaf expansion; Table 2 (Fig. 2).<sup>16</sup> More research is needed to determine the relationship between visible foliar injury and plant growth and/or yield.<sup>14</sup> At the present time there are no published experimental data linking realistic levels of acidic precipitation to visible injury to foliage or fruits that would reduce their marketability.

Loss of Forest Yield or Long-term Growth of Plants. There have been conflicting reports as to whether acidic precipitation can influence forest productivity. Tamm<sup>17</sup> concluded that except in areas where forest trees exhibit visible pollution symptoms, ambient acidic precipitation or other types of atmospheric acidity have no effect on tree growth. Other researchers found a statistically significant difference in tree growth between areas exposed to acidity and areas more remote from transported atmospheric acidity.<sup>10,18</sup> They concluded that there was no reason to attribute the growth reduction to anything other than acidic rain, but this by itself does not prove a cause-effect relationship. Researchers in Norway<sup>19</sup> and the United States<sup>20</sup> have not detected any consistent decreases in tree growth which could be attributed to acid deposition.

At the present time there has been no demonstration of losses of forest yield or forest productivity due to acidic precipitation. Experiments should be performed with adequate numbers of experimental replicates that could demonstrate economically significant differences between treatment means within 95% confidence limits. Experiments with adequate randomization of a large number of treatment replicates will avoid local soil and other micro-environmental problems that occur in agricultural fields and in natural terrestrial ecosystems.<sup>6</sup>

Alterations in photosynthesis and respiration could influence both forest and crop productivity. Photosynthesis and respiration in foliage may be influenced by exposure to acidic precipitation; however, research results are inconclusive. Leaves exposed to unrealistically low pH simulated rainfalls (pH 2.3) had lower sugar and starch contents compared with plants exposed to simulated rain of pH 5.7.<sup>11</sup> Foliage exposed to acidic rain may be stressed but short-term measurements of photosynthesis and respiration may not be sensitive enough to detect the degree of stress. Any relationship between the changes in carbohydrate status, on long- and short-term bases, with loss of plant and seed biomass, must be more firmly established before photosynthesis becomes a useful indicator of injury for the process of standard-setting.

Computer simulations of forest production using the JABAWA model, tested the effects of different levels of leaf area injury. Results showed that 10 to 20% per year of the leaf area of a tree species must be injured to change the role of that species or to reduce total forest biomass by a significant amount.<sup>21</sup> There are no indications that such extensive injuries do or will occur as a consequence of precipitation acidity.

Alterations in the nutrient content of foliage may influence the growth and productivity of both forests and crops. Moreover, nutrient levels in harvested portions of crops may affect the quality of foodstuffs. Acidic pre-

precipitation influences nutrient leaching from plant surfaces. Wood and Bormann<sup>22</sup> demonstrated that  $K^+$ ,  $Ca^{2+}$ , and  $Mg^{2+}$  were leached from leaves of pinto beans more rapidly at pH levels of 3.0 and 3.3 than at pH levels of 4.0 and 5.0.  $Ca^{2+}$  leached faster from foliage of sugar maple than  $K^+$  or  $Mg^{2+}$  at pH 3.0 than at pH 3.3. The leaching rates of  $K^+$  and  $Mg^{2+}$  were significantly higher at pH 3.0 than at pH 4.0. In tobacco leaves,  $Ca^{2+}$  leached faster from foliage exposed to simulated rain of pH 3.0 than from foliage exposed at pH 6.7.<sup>23</sup> In pinto beans exposed to simulated rain for 5 days, more calcium, nitrate, and sulfate were leached from foliage at pH levels of 2.7, 2.9, and 3.1 than at a control pH of 5.7.<sup>24</sup> In contrast, the amount of potassium leached was greater from leaves exposed to pH 5.7 than leaves exposed to pH levels between 3.4 and 2.9. The amounts of ammonium, magnesium, and zinc were the same at all pH levels tested. At the present time, insufficient experimental data are available to relate nutrient leaching and precipitation acidity with changes in vegetation growth or yield. As a result, nutrient leaching is not now a useful indicator of injury for the standard-setting process.

Acidic precipitation may change the surface characteristics of foliage. It has been suggested that acidic precipitation may effect the submicroscopic structure of the epicuticular wax layer(s) of leaves.<sup>25</sup> Some electron micrographs support this idea. This preferential erosion of the epicuticular waxes may be important for the penetration of rainwater. The effects of acidic precipitation on the cuticle are probably minimal, however. The most widely used methods of isolating cuticles involve exposure to strong acids.<sup>26,27</sup> Penetration rates of elements through isolated cuticles without epicuticular waxes were similar to rates through intact cuticles.<sup>27</sup> At the present time, there is no evidence that the leaf surface is altered by acidic precipitation to cause loss of crop or natural ecosystem productivity.

Nitrogen is, in general, the nutrient limiting productivity of temperate forests. Biological nitrogen fixation plus the deposition of ammonium and oxides of nitrogen are the routes by which this critical nutrient becomes available to forests. At Hubbard Brook, New Hampshire, bulk precipitation provides  $6.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , an amount equivalent to nearly 40% of the total nitrogen added to forest biomass annually.<sup>28</sup> The proportion of nitric acid in rain over the northeastern U.S. and in Colorado has been increasing and now accounts for one-third of the acidity in precipitation.<sup>29-32</sup> Thus there is a potential for positive effects of acidic precipitation on forests by fertilizing with nitrogen as well as the potential for injury due to acid effects on vegetation by inhibition of the nitrogen cycle and other microbial processes in soils, or increased leaching rates of nutrients from soils.

Computer simulation exercises were performed to test some of these effects. The JABAWA computer model of forest growth was modified to include effects of precipitation acidity on nitrogen availability and tree growth. The model found a direct positive relationship between N availability and biomass accumulation.<sup>33</sup> The relationships between forest production, litterfall, and decomposition of litterfall were analyzed using the FORTNIT forest nitrogen model. The simulation found lower rates of litter decomposition would be compensated by higher forest floor biomass accumulation so that N availabilities would not be greatly altered over a period of 15 to 40 years. After 40 years, slower litter decomposition rates provide higher N availabilities due to the decomposition of the larger litterfall biomass. However, no field data are available to confirm these simulations.<sup>33</sup>

Precipitation acidity may preferentially affect unicellular organisms that are very important in ecosystem function. Most multicellular organisms that have evolved from aqueous environments onto dry land have specialized surfaces to retard desiccation. Organisms with protective coverings may be less sensitive to acidic rain than organisms that lack these coverings.

Bacteria and other single-celled organisms are very sensitive to pH changes, presumably because they lack protective coverings. Procaryotic organisms seem to survive within a narrower range of pH levels than most multicellular organisms. Motility of most procaryotic organisms is greatest between pH 6.8 and 9.0.<sup>34-36</sup> Hoeninger<sup>37</sup> showed that a pH decrease results in an increase in abnormal wave motions in flagella of Proteus. Below pH 5.0 almost all motility ceased. The activities of bacteria are very important for the decomposition of leaf litter and thus the cycling of nutrients. However, although unicellular bacteria are very sensitive to the acidity in rainfall, with the present level of knowledge, the overall effects of natural levels of precipitation acidity on leaf decomposition and other related processes are unknown.

Visible Foliar Injury to Ornamental Plants that would Reduce Aesthetic Value. At the present time there is no evidence showing visible foliar injury to ornamental plants due to realistic precipitation acidities (excluding point source studies) that would reduce their market value. However, little research has been focused on this area.

Substantial Alterations of Plant Community Composition Leading to Ecosystem Simplification. If one or more plant species are preferentially affected by precipitation so they become greatly reduced or eliminated from plant communities, then the ecosystem may become simplified. This ecosystem simplification may eventually influence overall ecosystem productivity. Precipitation acidity could inhibit one or more critical stages in the life cycle of an organism. Experimental results show that fertilization and spermatozoid motility in ferns are very sensitive to pH levels below 5.7 and additions of sulfate and nitrate (43  $\mu\text{M}$ ).<sup>38-40</sup> These results suggest that the acidity of ambient precipitation in the northeastern United States inhibits the reproduction of ferns. The overall impact(s) that these injuries may have on ecosystem productivity is unknown.

Alterations in Herbivore Populations or Communities. It is conceivable that precipitation acidity might directly or indirectly alter populations of herbivores either positively or negatively, and such changes could be reflected in crop or forest productivity. However, at present, no data are available on the effects of precipitation acidity on terrestrial herbivores, let alone on subsequent impacts on plants.

Altered Sensitivity of Vegetation to Other Air Pollutants and/or Plant Pathogens. Acidic precipitation may affect gas exchange in plants which could alter plant sensitivity to other air pollutants. Individual cells of the epidermis may be injured upon initial exposure to acidic rain at pH levels of 4.0 and below.<sup>5,12-14</sup> Moreover, acidic rain may cause some alterations in the cuticle and/or in the functions of guard cells.<sup>24</sup> Foliage exposed to acidic rain may be more subject to wilting or water stress as well as becoming more sensitive to gaseous air pollutants. Knowledge of the effects of acidic rain in combination with gaseous pollutants such as ozone and sulfur dioxide is needed in order to understand the impacts precipitation acidity might have on vegetation in nature.

Acidic precipitation can affect microorganisms that inhabit surfaces of higher plants. Simulated acidic rain produced an 86% inhibition of the number of telia of Cronartium fusiforme on willow oak (Quercus phellos) and a 29% decrease in the percentage leaf area affected by bean rust (Uromyces phaseoli) on Phaseolus vulgaris.<sup>41</sup> Halo blight caused by Pseudomonas phaseolicola infections of leaves of Phaseolus vulgaris was stimulated or inhibited by simulated acidic rain depending upon the timing of application. Simulated acidic rain stimulated initial infection but inhibited development of the pathogen if applied after infection began. The overall effects that ambient levels of precipitation acidity have on plant host-plant pathogen interactions remain unknown at present. However, if rainfall acidity should increase or decrease,

the degree to which plant pathogens affect either crop or forest plants may have significant economic impacts.<sup>42,43</sup>

Overall Evaluation of Effects of Acidic Precipitation on Terrestrial Vegetation. At the present time there is little unequivocal evidence that acidic precipitation, at ambient levels, is having deleterious effects upon terrestrial vegetation within the United States. The above statement is made more from a lack of concrete information than from an evaluation of large amounts of experimental data. Experiments that have been conducted in laboratory or greenhouse settings have yielded some information that may point to mechanisms of injury. However, few field experiments have yielded definitive results. Although decreases in yields of various crops have been documented in controlled-environment and field experiments at pH levels below pH 4.0, only well-designed field experiments will document changes in plant productivity or survival that may be expected from actual acidic rain exposures. It is essential that experiments using standard agronomic practices with large numbers of replicates be conducted so that results can be extrapolated to actual crop values.

Acidic precipitation may affect the productivity of crop and forest plants by direct impaction. If significant alteration in productivity of either crops or forests occur, significant financial perturbations may result. Currently, evidence linking levels of rainfall acidity to crop yields is meager. However, from present data it is clear that the level of injury, if injury occurs, is less than year-to-year changes due to differences in natural climatic factors such as precipitation volume or temperature. This does not mean, however, that possible injuries from acidic precipitation can be ignored. It is important to determine even small changes in productivity. Modeling exercises show that a growth reduction of 10 to 20% per year of one tree species would be required to reduce overall forest productivity. Below this 10 to 25% range a decline in productivity of one or more species could be compensated for by an increased productivity of one or more other species.<sup>21,44</sup> However, it should be noted

that while such compensations may occur in complex ecosystems, they would not occur in agricultural systems. For example, a 2.6% reduction in yields (obtained in the experiment described above when plants were exposed to simulated rainfalls of pH 4.0)<sup>6</sup> of soybeans grown in the northeastern United States would represent a loss of 130 million dollars calculated for 1979 (ignoring price elasticity)<sup>45,46</sup>

In order to standardize results from future experiments, a uniform expression of data should be utilized. To date, the most meaningful relationships between rainfall acidity and plant responses have been obtained when the responses are plotted as a function of hydrogen ion concentration of the test solution applied.<sup>16,40</sup> This expression or some other more meaningful expression should be used so that data can be compared with ambient precipitation acidities.

Precipitation Acidity Standard for Protecting Forests and Crops. It has not yet been demonstrated that current regional levels of precipitation acidity alone significantly injure terrestrial vegetation (excepting areas affected by specific point sources). There are some suggestions that crop injury may be occurring, but the evidence is not conclusive. Since cause-effect relationships have not been shown, there is no information available on which to establish an acidity standard for precipitation that would prevent or ameliorate injuries. In the area of effects of acidic precipitation on terrestrial vegetation a much larger data base is required before a standard could be established, if indeed a standard is necessary.

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Table 1. Total protein content of seed samples of soybean plants exposed to simulated acidic rain.

Treatment (pH of simulated rainfalls)	Seed Mass plant <sup>-1</sup> (g) <sup>a/</sup>	Protein Content of seeds (mg mg <sup>-1</sup> ) <sup>b/</sup>	Seed Protein plant <sup>-1</sup> (g)	Reduction in protein content plant <sup>-1</sup> (%)
Ambient rain-				
falls only	19.4	0.324 <sup>x/c/</sup>	6.63	-
pH 4.0	18.9	0.316 <sup>y/</sup>	5.97	10.0
pH 3.1	18.1	0.296 <sup>y/</sup>	5.36	19.2
pH 2.7	17.2	0.298 <sup>y/</sup>	5.13	22.6
pH 2.3	17.8	0.302 <sup>y/</sup>	5.38	18.9

<sup>a/</sup> Data taken from Fig. 1

<sup>b/</sup> Protein analyses of seed samples were performed according to procedures of Thorne and Koller<sup>7</sup>. Total protein content was determined using the BIO-RAD Assay. Standards were made with freshly prepared bovine serum albumin. Analyses were performed on at least three samples from 10 randomly selected plots from each treatment.

<sup>c/</sup> Designates treatments where the protein content is significantly different ( $P < 0.05$ ) from the no-rain treatment. Numbers followed by + are not significantly different from each other. The Student-Newman-Keuls test<sup>8</sup> was applied to the data.

Table 2. Effect of simulated acidic rain on the rate of leaf elongation of pinto beans from day 2 to day 7.

Hydrogen ion concentration ( $\mu\text{eq l}^{-1}$ )	Leaf elongation rate <sup>a/</sup>	
	Mean Value	Significance
2	7.68	a <sup>b/</sup>
398	7.66	a
794	6.64	ab
1995	5.87	bc
3162	5.06	c

<sup>a/</sup> Leaflet elongation rate was calculated in  $\text{mm day}^{-1}$ . Rates are the line slopes from regression analyses. Regression analysis of leaflet elongation rates versus the hydrogen ion concentration of the applied rainfalls yielded a correlation coefficient of -0.98.

<sup>b/</sup> Means not followed by the same letter differ significantly ( $P < 0.05$ ) means followed by the same letter are not significantly different.

Fig. 1. Relationship between soybean seed yield per plant and the hydrogen ion concentration of the treatments. Ambient rainfalls provided an average hydrogen ion concentration of  $79.2 \mu\text{eq l}^{-1}$ . The four simulated acidic rainfall treatments provided hydrogen ion concentrations of 100, 794, 1995, and  $5012 \mu\text{eq l}^{-1}$  (pH levels of 4.0, 3.1, 2.7, and 2.3, respectively).

Fig. 2. Changes in leaflet length of middle leaflet of first trifoliate leaves of pinto beans (Phaseolus vulgaris) exposed to simulated rainfalls are shown. Rain acidity levels ( $\mu\text{eq l}^{-1} \text{H}^+$ ): 2, closed triangles; 398, squares, 794, open triangles; 1995, open circles; 3162, closed circles. Plants were 14 days old when the first, 6-min daily rainfall was applied. On day 0 leaves were about 40 mm in length. The largest leaflets attained a length of 110-120 mm after the ninth day of exposure. Straight lines were drawn to connect values from day 1 to day 2 and values from day 7 to day 9. Straight lines for values from day 2 to day 7 were drawn from regression analyses data. Statistical analyses of these data are shown in Table 2. The experiment was performed 4 times. Results of a typical experiment are shown.



