

## ASSESSMENT AND EVALUATION OF CRITICAL LEVELS FOR O<sub>3</sub> AND NH<sub>3</sub>

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### Abstract

The effect of O<sub>3</sub> on three different tree species was similar. A threshold of 40 ppb was found for *Pinus* and for *Pseudotsuga*. Over two growing seasons growth was inhibited in *Pinus* and *Fagus* saplings and total assimilation per tree was inhibited over one growing season in *Pseudotsuga* trees. *Pinus* was most sensitive to O<sub>3</sub>, but a critical level of 10 ppm.hrs is sufficient to protect the three species from O<sub>3</sub> damage. The effect of NH<sub>3</sub> on growth was ambiguous. Growth was unchanged in *Fagus*, stimulated in *Pseudotsuga* and inhibited in *Pinus*. The effect of NH<sub>3</sub> on tree architecture and stress sensitivity was similar. Tree architecture was changed in both *Fagus* and *Pseudotsuga* and drought and frost sensitivity were increased in *Pinus* and *Pseudotsuga*. At the moment both the critical level for O<sub>3</sub> and for NH<sub>3</sub> are exceeded in The Netherlands in general and in selected areas in particular.

### 1. INTRODUCTION

The current estimation of the mean nitrogen load in the Netherlands is 35 kg ha<sup>-1</sup> yr<sup>-1</sup> of which the largest proportion (16 kg ha<sup>-1</sup> yr<sup>-1</sup>) is gaseous ammonia. The regional mean NH<sub>3</sub> concentrations, however, may be very different from the average national concentration, resulting in a much higher exposure level for forest trees in areas with intensive live stock farming [1]. Ozone, another relevant air pollutant, is more evenly distributed over the Netherlands (Tab. 1). The national mean concentration exceeds the phytotoxic level for crops [2], and the economic impact on Dutch crops is large [3]. The effect of O<sub>3</sub> on mature forest trees, on forest ecosystems and on natural ecosystems in general is largely unknown. The available information is mainly acquired from experiments with seedlings and saplings [4].

Two types of air quality standards are currently in use: critical levels and critical loads. Critical levels are based on exposure values, while critical loads are based on deposition values. The use of two different air quality standards is justified, because critical levels focus on individual air pollution components and on non-depositing oxidants such as O<sub>3</sub>, whereas critical loads lump either nitrogen or

acidifying components together. Moreover critical levels are usually used for short-term exposures (hours - months) and critical loads for long-term exposures (years).

The aim of the present investigation was to assess the impact of  $\text{NH}_3$  and  $\text{O}_3$  on forest ecosystems at concentrations presently occurring in the Netherlands in relation to the critical level. This was done by comparing the effects on tree saplings exposed to both pollutants in OTC's (Open Top Chambers) with the effects on mature forest trees under ambient field conditions.

## 2. MATERIAL AND METHODS

### 2.1 Fumigation experiments in OTC's

Two experiments were performed in OTC's, in which 3-yr-old Beech (*Fagus sylvatica* L.) and Scots pine (*Pinus sylvestris* L.) saplings were fumigated for 15 months, largely covering two growing seasons (from June 1 to September 1). The OTC's have been described earlier [5]. Ammonia and ozone were injected into the air stream prior to the blower via thermal mass-flow controllers (Brooks 5850 TR). Air pollutant concentrations were sequentially monitored with an ozone analyzer (8810, Monitor Labs) and a  $\text{NH}_3$  monitor (thermoconverter model 8750 followed by a chemiluminescent  $\text{NO}_x$  analyzer model 8840, both Monitor Labs) and were computer-controlled. The data were recorded with a HP data acquisition system. A duplicated range of  $\text{O}_3$  concentrations (0, 30, 60, 90, 120 and  $150 \mu\text{g m}^{-3}$ ) was used in the first experiment and these concentrations were both higher and lower than current ambient concentrations in the Netherlands (Tab. 1).

Table 1  
Average concentrations of  $\text{NH}_3$  and  $\text{O}_3$  ( $\mu\text{g m}^{-3}$ ) in 1992.

	$\text{NH}_3$	$\text{O}_3$
The Netherlands	3.4	60
Wageningen, OTC's	16	72
Veluwe, field	2.5	78

Trees were exposed to  $\text{O}_3$  during a 9 h day and to a third of the daytime concentrations during the remaining 15 h. To one of the  $\text{O}_3$  ranges, a concentration of  $40 \mu\text{g m}^{-3}$   $\text{NH}_3$  was added ( $24 \text{ h day}^{-1}$ ), which is somewhat higher than the highest mean concentrations experienced in the Netherlands. In the control treatments, the  $\text{NH}_3$  concentration was also higher ( $15 \mu\text{g m}^{-3}$ ) than the national mean due to the fact that Wageningen is located in a region with high  $\text{NH}_3$  concentrations (Tab. 1), and the filters used to clean the air have less than 50% capacity for  $\text{NH}_3$ . In the second experiment,  $\text{O}_3$  and  $\text{NH}_3$  were applied in factorial design, resulting in a triplicated set of  $\text{O}_3$  concentrations (setpoints 0, 90, and  $135 \mu\text{g m}^{-3}$ ), supplemented with ambient air, or with  $\text{NH}_3$  to  $40 \mu\text{g m}^{-3}$  and  $80 \mu\text{g m}^{-3}$   $\text{NH}_3$ . The water potential on 1-year-old

needles of *Pinus sylvestris* was measured with a pressure bomb [6]. The measurements were performed in six OTC's only, in all three NH<sub>3</sub> treatments combined with filtered air and the highest O<sub>3</sub> concentration and were performed between 08.00 and 13.00 hours in fully watered pots and again after five days without water. The soil water potential was measured daily to ensure that the drought treatment was not prolonged to the point where excessive drought injury to the trees occurred in order to relate the soil water potential to the needle water status. This paper discusses the main results on biomass production and drought sensitivity.

## 2.2 Correlation studies in the field

All field measurements were performed in a stand of 34-year-old Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco L., provenance Arlington), located at the Veluwe, in the central part of the Netherlands. The Speuld site has a stand density of approximately 800. In 1993, the average tree height was 22.2 m and the average DBH was 25.4 cm [7]. The mean O<sub>3</sub> and NH<sub>3</sub> concentration at Speuld are shown in Tab. 1. A computer controlled field gas exchange system was installed at the site. With this system up to 16 different branch assimilation chambers can be measured continuously. A 22 m high scaffolding was built in the middle of the stand, from which the branch assimilation chambers could be mounted in 8 different trees. The chambers were ventilated with ambient air and contained one year class of needles. Temperature, relative humidity and CO<sub>2</sub> concentration of the air entering the chambers closely resembled that of ambient outside air. A PAR sensor was mounted at the outside of each chamber. Each chamber was sampled twice every hour during 24 hours per day from March 1992 up to December 1993. The light response curve was modelled using the equation of Goudriaan [8] with the measured photosynthetic rate and photoactive radiation levels. The unexplained variance in the data could be reduced by 40% by taking into account changes in vapour pressure deficit (VPD) and the O<sub>3</sub> concentration. In this way, the direct effects of O<sub>3</sub>, NH<sub>3</sub> and NO<sub>x</sub> and VPD on the photosynthetic rate were statistically estimated. This paper discusses the results on CO<sub>2</sub> assimilation, per month and per year, and on the needle nutrient status.

## 2.3 Exposure-response relationship for O<sub>3</sub>

The levels of exposure to O<sub>3</sub> used here are not expressed as concentrations, but as the *accumulated exposure over a threshold concentration*, abbreviated with AOT [4]. The AOT adds all exposures above a certain threshold concentration over the period of interest, i.e. growing season, but also over several years in which mean concentrations can strongly fluctuate over the seasons. The threshold value is the concentration above which O<sub>3</sub> toxicity becomes evident. The AOT approach proved to be very useful for evaluation of crop loss in which relative yield reductions could be linearly fitted to AOT values. Recently, this approach has also been applied to forest trees, although reliable field data are scarce [4].

### 3. RESULTS

#### 3.1. Fumigation experiments in OTC's

Fig. 1 shows the effects of  $O_3$  and  $NH_3$  on the total biomass of *P. sylvestris*. The biomass of trees exposed to the lowest 3 levels of  $O_3$  was significantly higher than that in the 3 highest levels. An effect threshold for  $O_3$  is visible between 5 and 15 ppm-hrs. The 3 lowest  $O_3$  concentrations below 40 ppb were logically similar in terms of AOT40, and this was also reflected in the biomass production in those treatments, indicating the validity of the threshold for *P. sylvestris*. The absence of a linear exposure-response relationship is likely due to the fact that high concentrations of  $O_3$  inhibit stomatal conductance [9] and thus the uptake of  $O_3$ . The UNECE and the WHO recently recommended an AOT40 of 10 ppm-hrs as a critical level for trees, at which a 10% reduction in biomass production should not be exceeded. The results shown for *P. sylvestris* in Fig. 1 appear to support this critical level. Current concentrations of  $O_3$  in the Netherlands however, exceed this critical level by more than a factor 2, indicating that  $O_3$  is significantly reducing the growth of *P. sylvestris*. Fig. 1 also indicates that at low  $O_3$  concentration,  $NH_3$  tends to inhibit growth, but growth is not further reduced at higher  $O_3$  concentrations.

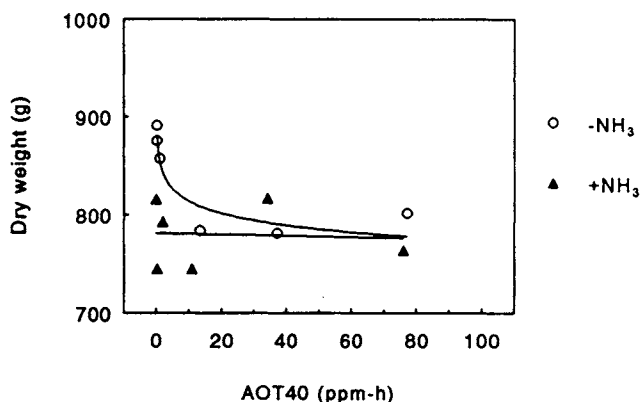


Figure 1. Effect of  $O_3$  and  $NH_3$  on the total biomass production *Pinus sylvestris*.  $\circ$  =  $O_3$  alone;  $\blacktriangle$  =  $O_3 + NH_3$ .

In the same fumigation experiment, *F. sylvatica* was found to be less sensitive to  $O_3$ ; a 10% reduction in biomass production was found at an AOT40 of 30 ppm-hrs, while  $NH_3$  had no effect on the total biomass. The growth of *F. sylvatica* was differentially affected by  $NH_3$  and  $O_3$  in tree architecture rather than in biomass production (Fig. 2). Although  $O_3$  inhibited tree height, it increased stem diameter, which resulted in relatively sturdier trees.  $NH_3$  did not influence tree height and reduced stem diameter, resulting in relatively smaller trees at higher levels of  $O_3$ . Lateral branch growth was also reduced by  $NH_3$  with increasing concentrations of  $O_3$ , thus reducing the potential for light interception.

In the second fumigation experiment, special attention was paid to the effect of  $NH_3$  and  $O_3$  on the drought sensitivity of *P. sylvestris*. The data in Fig. 3 show that needles of fully watered *P. sylvestris* saplings have a significantly higher (more negative) water potential when exposed to  $NH_3$  alone than when exposed to  $NH_3 + O_3$ .

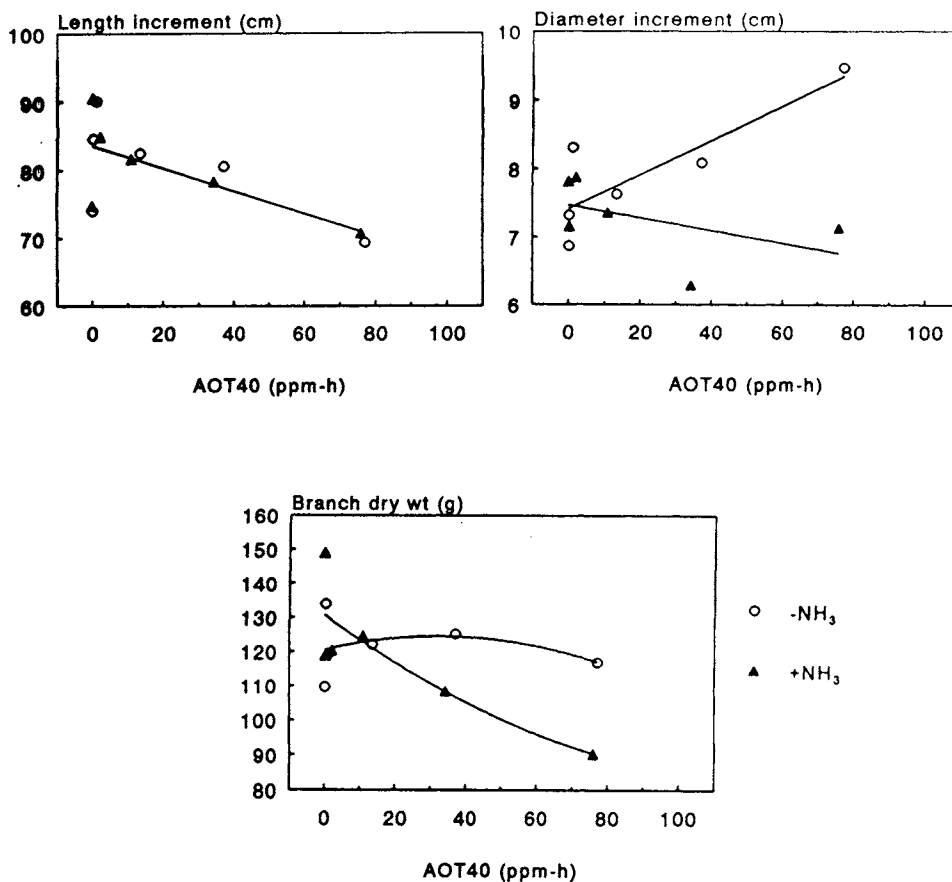


Figure 2. Effect of  $O_3$  and  $NH_3$  on mean length increment, stem diameter increment and branch dry weight of *Fagus sylvatica*.  $\circ$  =  $O_3$  alone;  $\blacktriangle$  =  $O_3 + NH_3$ .

No difference was observed in the needle water potentials between the three treatments of only  $NH_3$ . It appears that, irrespective of the  $NH_3$  concentration,  $O_3$  inhibits stomatal conductance and thus transpiration in the absence of water stress, allowing the trees to maintain turgor at a lower (200-400 kPa) needle water potential than when exposed to  $NH_3$  alone. When the trees were droughted for 5 days, the water potential increased linearly with increasing concentrations of  $NH_3$  in the absence of  $O_3$ . This indicates that under conditions of drought stress,  $NH_3$  disrupts the stomatal control, resulting in increased transpiration and reduced water use efficiency [10-11]. When  $O_3$  was added to the  $NH_3$  treatments, the effect of increasing  $NH_3$  concentrations was masked, and the water potential in all three  $O_3 + NH_3$  treatments remained lower than in treatments with  $NH_3$  alone. This confirms earlier experiments, in which the reduced transpiration of various species exposed to  $O_3$  is attributed to inhibition of stomatal aperture [9]. Thus, even at the concentrations of  $NH_3$  used in this experiment,  $O_3$  appears able to reduce water loss through transpiration and is thus able to reduce drought stress.

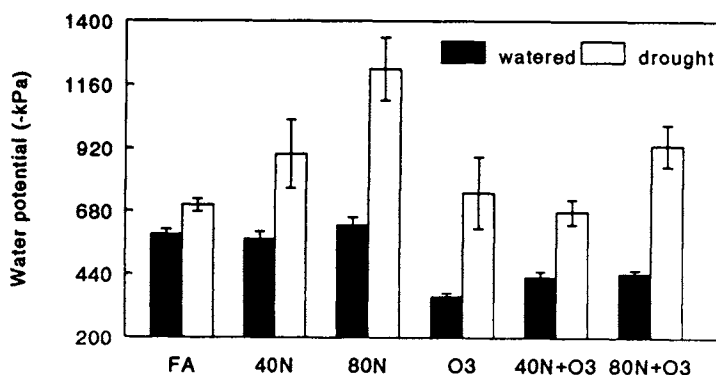


Figure 3. Effect of O<sub>3</sub> and NH<sub>3</sub> on drought stress of *Pinus sylvestris*. Dark bars indicate water potential (means  $\pm$  SE) of watered trees and white bars that of droughted trees (n=10).

### 3.2. Correlation studies in the field

The vitality of the Speuld stand was characterized in 1986 according to international EC standards of defoliation and foliar discoloration. The vitality was higher than the nationwide average for *P. menziesii*, which was classified as less healthy for 50% of all trees [12]. The vitality was re-assessed in 1994, and was similar to the nationwide average, indicating that the vitality of the stand had decreased to the level of most other *P. menziesii* stands in The Netherlands, which was classified as less healthy for 80% of all trees [7].

Notwithstanding the poor vitality, biomass production was high. According to yield tables for Dutch *P. menziesii* stands [13], based on data from the pre-intensive livestock age, annual wood volume increment was higher than expected. Speuld can be qualified as an average soil quality site (class III) with an expected mean annual volume increment of around 13.4 m<sup>3</sup> ha<sup>-1</sup>. The average annual increment over the period 1987-1993 however, was 24.7 m<sup>3</sup> ha<sup>-1</sup>, which is even higher than the expected volume increment of 17.3 m<sup>3</sup> ha<sup>-1</sup> on the best quality site class.

Table 2. Biomass partitioning in *Pseudotsuga menziesii* using foliage [7] and root [14] data from Speuld and from [15-16].

	Speuld	low productive, slow grow rate	high productive, fast grow rate
Foliage/ fine root	5.5	0.95-1.1	2.7-3.6
Foliage/ coarse root	0.67	0.21-0.34	0.19-0.31

The dry weight distribution in Speuld in which the N deposition is lower than the Dutch mean, was compared to that of other *P. menziesii* stands. Speuld can be best compared to high productive, fast growing stands of the same age. The foliage/root ratio in Speuld is exceptionally high compared to the other stands (Tab. 2). The high foliage/root ratio results from both lower root biomass and higher needle biomass. The high amount of needle biomass is also reflected in the LAI of the stand which was 10.7 in 1992. The foliage/coarse root ratio is also high (Tab. 2). The branch dry weight however, was lower compared to the other stands.

The nutrient status of the needles changed significantly during the experimental period [8]. The average N concentration increased from 1.7% in 1987 to 2% in 1993. The optimal concentration for biomass production in *P. menziesii* is 1.8%. The K concentration decreased from 0.7% to 0.5%, which is below the deficiency level of 0.6%. The P concentration remained constant, but was also below the deficiency level of 0.14%. The N/P ratio was constantly below/above the deficiency level, while the N/K ratio increased from 2.7 to 3.7. Extrapolating the nutrient trends found over the past 7 years, the ratio of N/K can be expected to reach the deficiency level within 1-2 years. High N/K ratio's, as found in the Speuld stand, are often considered as an indicator for increased stress sensitivity, e.g. frost sensitivity [17]. There seems to be little doubt that the high nitrogen status, the high productivity, the high foliage/root ratio, the high LAI and the increased stress sensitivity are caused by the high N deposition into the stand.

Table 3. Proportional reduction of the monthly CO<sub>2</sub> assimilation due to O<sub>3</sub> and VPD (mean over 2 needle age classes and three crown levels).

	O <sub>3</sub>	VPD	O <sub>3</sub>	VPD
	1992		1993	
April	3.3	2.4	28.0	14.0
May	12.2	22.9	13.2	8.1
June	4.4	20.1	4.4	6.1
July	5.3	6.3	7.6	2.9
August	4.1	7.0	5.4	NS

Net CO<sub>2</sub> assimilation was measured more or less constantly throughout 1992 and 1993 and was related to changes in meteorological parameters and air pollution concentration. No direct effects of NH<sub>3</sub> and/or NO<sub>x</sub> on net CO<sub>2</sub> assimilation of *P. menziesii* were found. NH<sub>3</sub> tended to stimulate assimilation, but the differences were not significant. The only obvious relationship of CO<sub>2</sub> assimilation with air pollution was that with O<sub>3</sub> (Tab. 3). The reduction of net CO<sub>2</sub> assimilation by O<sub>3</sub> on a monthly basis can be quite considerable, and was in the same range as the reduction by VPD.

The annual reduction of total CO<sub>2</sub> assimilation per tree was estimated to be in the range of 3-10% in 1992. The annual reduction over 1993 will probably be higher, as the monthly reductions are higher. In order to assess the critical level for O<sub>3</sub> for adult *P. menziesii* trees under ambient conditions, the reduction in biomass production by O<sub>3</sub> has to be known. We assumed that the annual reduction in total CO<sub>2</sub> assimilation is in the same range as the annual reduction in biomass production. Matyssek et al. however, (pers. commun.) found that a 40% reduction in total CO<sub>2</sub> assimilation resulted in a 60% reduction in biomass.

#### 4. EVALUATION AND CONCLUSIONS

Despite the fact that different tree species were used, the reaction of saplings and adult trees under field conditions to high NH<sub>3</sub> concentrations showed both similarities and discrepancies. Most of the work done on effects of NH<sub>3</sub> on plants to date indicate that NH<sub>3</sub> stimulates growth, but the results presented in this paper show that NH<sub>3</sub> may also inhibit growth in the presence of low concentrations of O<sub>3</sub> in *Pinus*. In *Fagus* however, NH<sub>3</sub> had no effect on growth, but changed tree architecture by decreasing tree height and branch biomass. Under field conditions the high N input resulted in both increased growth and in a changed architecture. Dry weight distribution in *Pseudotsuga* was affected, as foliar biomass was higher and fine root and branch biomass was lower. Another similarity was the increased sensitivity. NH<sub>3</sub> was found to increase drought sensitivity in *Pinus* saplings, and both drought and frost sensitivity in adult *Pseudotsuga* trees under ambient conditions. NH<sub>3</sub> is the major part of the total N deposition in the Netherlands. The aim to reduce emissions until critical loads are not exceeded, will surely reduce NH<sub>3</sub> levels below the critical level for NH<sub>3</sub>. The critical level proposed for NH<sub>3</sub> is 8 and 270 µg m<sup>-3</sup> respectively, for an annual and a 24 hrs mean [18]. The annual mean is currently being exceeded in half of the Dutch areal [1]. On sites removed from the direct influence of point sources, the 24 h critical level is probably not being exceeded, not even in the Netherlands. Exceedances are frequent only on a local scale, in the first 300 meters from a point source. If environmental policy will be based on damage estimates in relation to emission and dispersion, knowledge of effect thresholds of NH<sub>3</sub> is insufficient. Although NO<sub>x</sub> is only a small part of the total N deposition, its abatement would be very profitable because it would reduce the O<sub>3</sub> concentration as well.

High O<sub>3</sub> concentrations resulted in decreased growth in all tree species. However, a 10% reduction in biomass production was reached at different AOT40 values (Fig. 4). It seems that the critical level for forest trees of 10 ppm.hrs is able to protect our trees from damage by O<sub>3</sub>. From the range of AOT40 values between '87-'93 it is evident that these values are exceeded in general and in the forested area of the Veluwe in particular. Timber producers may not bother about a 10% reduction in biomass by O<sub>3</sub> if the fertilizing effect of atmospheric N deposition compensates for the losses by O<sub>3</sub>. However, this does not hold for the Dutch situation. First of all growth stimulation by N is of a temporary nature. An initial growth stimulation by N is accompanied by increasing ratio's of N to cations and following luxurious N consumption will eventually lead to a decreased growth due to severe cation deficiencies [19]. The progressing nutrient status of Speuld suggests that it will not



take decades to reach this situation. Furthermore, a number of forested areas in The Netherlands appears to have reached the stage of growth reduction already [19].

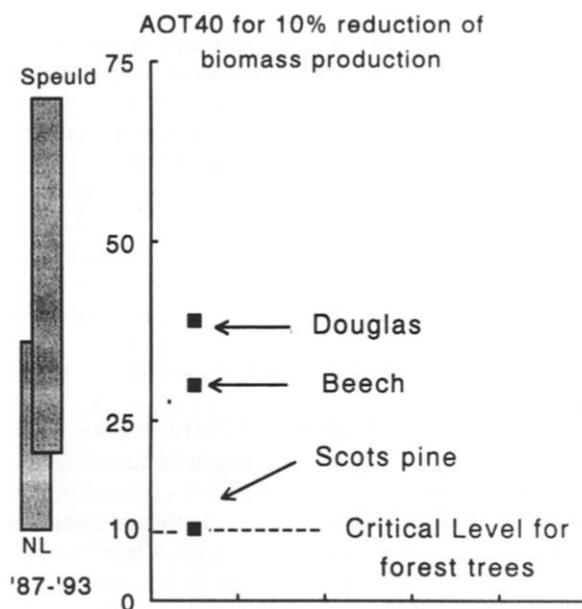


Figure 4. AOT40 values for forest trees. The columns on the left indicate the range of O<sub>3</sub> concentrations measured during 1987-1993 in Speuld (25m height) and as a national average (3m). The arrows indicate the AOT40 values for *Pinus sylvestris*, *Fagus sylvatica* saplings and for mature *Pseudotsuga menziesii*, presented in this paper in relation to the critical level proposed for forest trees.

Secondly, forests are more than trees alone. The impact of air pollution on trees (and crops) should not be seen as indicative of the unknown effects on other parts of the forest ecosystem. Effects on herbs and grasses in the undergrowth and on biodiversity of plants and animals are likely to be more pronounced [20]. The results presented in this paper suggest that all three tree species used in the experiments are adversely affected by current O<sub>3</sub> concentrations in The Netherlands. The possibilities to quantify this damage has increased strongly in recent years, but much has yet to be done.

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