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equilibrium model for downregulation and the reversible radical pair

Kevin Oxborough and Neil R. Baker

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 An evaluation of the potential triggers of photoinactivation of photosystem II An evaluation of the potential triggers
of photoinactivation of photosystem II
in the context of a Stern–Volmer model
for downroquistion and the reversible of photoinactivation of photosystem II
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for downregulation and the reversible
redical poir equilibrium model **e context of a Stern–Volmer mo
downregulation and the reversik
radical pair equilibrium model**

Kevin Oxborough* **and Neil R. Baker**

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Photoinactivation of photosystem II (PS II) is a light-dependent process that frequently leads to break-Photoinactivation of photosystem II (PS II) is a light-dependent process that frequently leads to break-
down and replacement of the D1 polypeptide. Photoinhibition occurs when the rate of photoinactivation
is greater than Photoinactivation of photosystem II (PS II) is a light-dependent process that frequently leads to break-
down and replacement of the D1 polypeptide. Photoinhibition occurs when the rate of photoinactivation
is greater than down and replacement of the DI polypeptide. Photoinhibition occurs when the rate of photoinactivation
is greater than the rate at which DI is replaced and results in a decrease in the maximum efficiency of PS
II photochemi is greater than the rate at which D1 is replaced and results in a decrease in the maximum efficiency of PS II photochemistry. Downregulation, which increases non-radiative decay within PS II, also decreases the maximum eff II photochemistry. Downregulation, which increases non-radiative decay within PS II, also decreases the maximum efficiency of PS II photochemistry and plays an important role in protecting against photoinhibition by reduci maximum efficiency of PS II photochemistry and plays an important role in protecting against photoinhi-
bition by reducing the yield of photoinactivation. The yield of photoinactivation has been shown to be
relatively inse bition by reducing the yield of photoinactivation. The yield of photoinactivation has been shown to be
relatively insensitive to photosynthetically active photon flux density (PPFD). Formation of the P680
radical (P680⁺) relatively insensitive to photosynthetically active photon flux density (PPFD). Formation of the P680
radical (P680⁺), through charge separation at PS II, generation of triplet-state P680 (³P680^{*}), through
intersyst radical (P680⁺), through charge separation at PS II, generation of triplet-state P680 (³P680^{*}), through intersystem crossing and charge recombination, and double reduction of the primary stable electron acceptor of intersystem crossing and charge recombination, and double reduction of the primary stable electron acceptor of PS II (the plastoquinone, Q_A) are all potentially critical steps in the triggering of photoinactivation. In acceptor of PS II (the plastoquinone, Q_A) are all potentially critical steps in the triggering of photoinactivation. In this paper, these processes are assessed using fluorescence data from attached leaves of higher pla vation. In this paper, these processes are assessed using fluorescence data from attached leaves of higher
plant species, in the context of a Stern–Volmer model for downregulation and the reversible radical pair
equilibriu plant species, in the context of a Stern–Volmer model for downregulation and the reversible radical pair
equilibrium model. It is shown that the yield of P680⁺ is very sensitive to PPFD and that downregulation
has very l equilibrium model. It is shown that the yield of $P680^+$ is very sensitive to $PPFD$ and that downregulation has very little effect on its production. Consequently, it is unlikely to be the trigger for photoinactivation. has very little effect on its production. Consequently, it is unlikely to be the trigger for photoinactivation.
The yields of ³P680^{*} generated through charge recombination or intersystem crossing are both less sensiti tive to PPFD than the yield of P680⁺ and are both decreased by downregulation. The yield of doubly tive to PPFD than the yield of P680⁺ and are both decreased by downregulation. The yield of doubly reduced Q_A increases with incident photon flux density at low levels, but is relatively insensitive at moderate to hig reduced Q_A increases with incident photon flux density at moderate to high levels, and is greatly decreased by downreg reduced Q_A are both viable as triggers of photoinactivation. reduced Q_A are both viable as triggers of photoinactivation.
Keywords: photosynthesis; photoinhibition; chlorophyll fluorescence

1. INTRODUCTION

1. INTRODUCTION
The well-documented, extreme vulnerability of photo-
system II (PS II) to light-induced damage is almost **SET ATTRODUCTION**
System II (PS II) to light-induced damage is almost
certainly linked to its unique ability to oxidize water The well-documented, extreme vulnerability of photosystem II (PS II) to light-induced damage is almost
certainly linked to its unique ability to oxidize water.
Oxygenic organisms are able to overcome this vulnersystem II (PS II) to light-induced damage is almost
certainly linked to its unique ability to oxidize water.
Oxygenic organisms are able to overcome this vulner-
ability through the rapid turnover of what would appear certainly linked to its unique ability to oxidize water.
Oxygenic organisms are able to overcome this vulner-
ability through the rapid turnover of what would appear
to be the main target of this damage, the Dl polypentide Oxygenic organisms are able to overcome this vulner-
ability through the rapid turnover of what would appear
to be the main target of this damage, the D1 polypeptide
of PS II. Prior to breakdown of D1. PS II is 'photoinact ability through the rapid turnover of what would appear
to be the main target of this damage, the D1 polypeptide
of PS II. Prior to breakdown of D1, PS II is 'photoinacti-
vated' through the action of one or more light-dep to be the main target of this damage, the DI polypeptide
of PS II. Prior to breakdown of DI, PS II is 'photoinacti-
vated' through the action of one or more light-dependent
mechanisms. In this study, potential triggers of of PS II. Prior to breakdown of Dl, PS II is 'photoinactivated' through the action of one or more light-dependent mechanisms. In this study, potential triggers of photoinacvated' through the action of one or more light-dependent
mechanisms. In this study, potential triggers of photoinac-
tivation are evaluated, taking into account the various
pathways for dissination of absorbed excitation e mechanisms. In this study, potential triggers of photoinactivation are evaluated, taking into account the various pathways for dissipation of absorbed excitation energy at PS II tivation
pathway
PS II.
The I pathways for dissipation of absorbed excitation energy at PS II.
The D1 polypeptide forms one-half of the heterodimeric

core of PS II, which binds all of the major redox components involved in charge separation and stabilization (the core of PS II, which binds all of the major redox components involved in charge separation and stabilization (the other half being the D2 polypeptide, which is also replaced in some situations). Charge separation at PS II nents involved in charge separation and stabilization (the other half being the D2 polypeptide, which is also replaced in some situations). Charge separation at PS II replaced in some situations). Charge separation at PS II
*Author for correspondence (koxbor@essex.ac.uk).

leads to formation of the radical pair, $P680⁺/Phe⁻$ (see leads to formation of the radical pair, $P680^+/Phe^-$ (see
Appendix A for explanation of abbreviations). $P680^+$ has leads to formation of the radical pair, $P680^+/Phe^-$ (see
Appendix A for explanation of abbreviations). $P680^+$ has
a redox potential that is high enough (1V or more), not
only to oxidize water, but also the nigment molec Appendix A for explanation of abbreviations). $P680^+$ has
a redox potential that is high enough (1V or more), not
only to oxidize water, but also the pigment molecules of
 $PS II$ (chlorophyll a and β -carotene) or possibl a redox potential that is high enough $(1V \text{ or more})$, not only to oxidize water, but also the pigment molecules of only to oxidize water, but also the pigment molecules of PS II (chlorophyll *a* and β -carotene) or possibly the Dl protein itself (Anderson *et al.* 1998). Consequently, there is PS II (chlorophyll *a* and β -carotene) or possibly the Dl e) or possibly the DI
Consequently, there is
is a direct trigger of photoinactivation. a very real possibility that $P680^+$ is a direct trigger of photoinactivation.
Exciton transfer among chlorophylls within the light-

harvesting system associated with PS II can lead to formation of the triplet excited state $(^{3}ChI^{*})$ from the singlet excited state $(^1Chl^*)$ through intersystem crossing. formation of the triplet excited state $(^{3}Ch)^{*}$) from the singlet excited state $(^{1}Ch)^{*}$) through intersystem crossing.
Although ³Chl^{*} is not likely to induce damage directly, quenching of ³Chl^{*} by triplet ove singlet excited state (¹Chl^{*}) through intersystem crossing.
Although ³Chl^{*} is not likely to induce damage directly,
quenching of ³Chl^{*} by triplet oxygen (³O₂) can result in
formation of singlet excited oxy Although ³Chl^{*} is not likely to induce damage directly,
quenching of ³Chl^{*} by triplet oxygen (³O₂) can result in
formation of singlet excited oxygen (¹O₂^{*}), which has been
shown to result in preferential quenching of ³Chl^{*} by triplet oxygen (³O₂) can result in formation of singlet excited oxygen (¹O₂^{*}), which has been shown to result in preferential destruction of P680 and formation of singlet excited oxygen $(^1O_2^*)$, which has been
shown to result in preferential destruction of P680 and
D1 breakdown (Shipton & Barber 1991; Vass *et al.* 1992).
In aerobic photosynthetic organisms, interact shown to result in preferential destruction of P680 and
DI breakdown (Shipton & Barber 1991; Vass *et al.* 1992).
In aerobic photosynthetic organisms, interaction between
singlet ground-state B-carotene (¹Car) and ³Chl DI breakdown (Shipton & Barber 1991; Vass *et al.* 1992).
In aerobic photosynthetic organisms, interaction between
singlet ground-state β -carotene (¹Car) and ³Chl^{*} results
in formation of the triplet excited stat In aerobic photosynthetic organisms, interaction between
singlet ground-state β -carotene (¹Car) and ³Chl^{*} results
in formation of the triplet excited state of β -carotene
(³Car^{*}) and ¹Chl effectively redu $(^{3}Car^{*})$ and 'Chl, effectively reducing the lifetime of the inglet ground-state β-carotene (¹Car) and ³Chl^{*} results
n formation of the triplet excited state of β-carotene
³Car^{*}) and ¹Chl, effectively reducing the lifetime of the

³Chl* by several orders of magnitude (Siefermann-Harms 3 Chl^{*} by several orders of magnitude (Siefermann-Harms & Angerhofer 1998, and references therein). The 3 Car^{*}, formed through this interaction, then reverts to the 3 Chl $*$ by several orders of magnitude (Siefermann-Harms & Angerhofer 1998, and references therein). The 3 Car $*$, formed through this interaction, then reverts to the singlet ground state through intersystem cro & Angerhofer 1998, and references therein). The ³Car^{*},
formed through this interaction, then reverts to the
singlet ground state through intersystem crossing
 β Car^{*}→Car) 8-carotene provides an additional level of $(3\text{Car}^* \rightarrow 1\text{Car})$. β-carotene provides an additional level of bormed through this interaction, then reverts to the
inglet ground state through intersystem crossing
³Car^{*} → ¹Car). β-carotene provides an additional level of
protection against ¹O^{*} through direct interaction. singlet ground state through intersystem crossing $({}^{3}$ Car^{*} \rightarrow ¹Car). β-carotene provides an additional level of protection against 1 O₂^{*} through direct interaction. This leads to formation of 3 O_c and $\binom{3}{2}$ Car^{*} \rightarrow ¹Car). β-carotene provides an additional level of protection against ¹O₂^{*} through direct interaction. This leads to formation of ³O₂ and ³Car^{*} and decreases the lifetime of any ¹O^{*} protection against ${}^{1}O_{2}^{*}$ through direct interaction. This
leads to formation of ${}^{3}O_{2}$ and ${}^{3}Car^{*}$ and decreases the
lifetime of any ${}^{1}O_{2}^{*}$ that may form through interaction
with 'unquenched' ${}^{3}Ch1$ leads to formation of ${}^{3}O_{2}$
lifetime of any ${}^{1}O_{2}^{*}$ that if
with 'unquenched' ${}^{3}Chl^*$.
The distribution of spect etime of any ¹O₂^{*} that may form through interaction
th'unquenched'³Ch1^{*}.
The distribution of spectral types of chlorophylls *a* and
within the light-harvesting system of PS II results in a

with 'unquenched' ³Chl^{*}.
The distribution of spectral types of chlorophylls *a* and cally active photon flux density (PPFD).
b within the light-harvesting system of PS II results in a There are two processes operating higher density of excitation energy at and around the reaction centre (Dau & Sauer 1996). A consequence of higher density of excitation energy at and around the reaction centre (Dau & Sauer 1996). A consequence of this is that the yield of 3 Chl^{*} formation is highest at the core of the PS II light-harvesting system, whic reaction centre (Dau & Sauer 1996). A consequence of
this is that the yield of 3 Chl^{*} formation is highest at the
core of the PS II light-harvesting system, which includes
 P_{max} Evidence has recently been presen this is that the yield of 3 Chl^{*} formation is highest at the core of the PS II light-harvesting system, which includes P_{680} . Evidence has recently been presented that the macrostructure of the region of the ligh core of the PS II light-harvesting system, which includes P_{680} . Evidence has recently been presented that the macrostructure of the region of the light-harvesting system closest to the reaction centre acts as a harrie P_{680} . Evidence has recently been presented that the macrostructure of the region of the light-harvesting system closest to the reaction centre acts as a barrier to the diffusion of Ω , thereby providing yet another macrostructure of the region of the light-harvesting
system closest to the reaction centre acts as a barrier to
the diffusion of O_2 , thereby providing yet another level of
protection against ${}^1O^*$ formation where th system closest to the reaction centre acts as a barrier to
the diffusion of O_2 , thereby providing yet another level of
protection against ${}^1O_2^*$ formation where the yield of ³Chl^{*}
formation is likely to be high the diffusion of O_2 , thereby providing yet another level of
protection against ${}^1O_2^*$ formation where the yield of ${}^3Chl^*$ 19
formation is likely to be highest (Siefermann-Harms & a protection against ¹
formation is likely
Angerhofer 1998).

Angerhofer 1998).
As noted above, the oxidizing potential of P_{680}^+ is high
enough to cause irreversible oxidation of β -carotene and/
or adiacent chlorophylls. Consequently, although there is As noted above, the oxidizing potential of P_{680}^+ is high
enough to cause irreversible oxidation of β -carotene and/
or adjacent chlorophylls. Consequently, although there is
at least one β -carotene within the co enough to cause irreversible oxidation of β -carotene and/
or adjacent chlorophylls. Consequently, although there is
at least one β -carotene within the core complex of PS II,
there is some doubt as to whether or not or adjacent chlorophylls. Consequently, although there is
at least one β -carotene within the core complex of PS II,
there is some doubt as to whether or not it could be
located close enough to P680 to be able to quench at least one β -carotene within the core complex of PS II,
there is some doubt as to whether or not it could be
located close enough to P680 to be able to quench any
 ${}^{3}P_{\infty}^{*}$, that may be formed through intersyst ${}^{3}P_{680}^{*}$ that may be formed through intersystem crossing or there is some doubt as to whether or not it could be located close enough to P680 to be able to quench any located close enough to P680 to be able to quench any
 ${}^{3}P_{680}^{*}$ that may be formed through intersystem crossing or
as the result of charge recombination between Phe⁻ and
 P^{+} (Barber 1998) Consequently P^{+} ma ${}^{3}P_{680}^{*}$ that may be formed through intersystem crossing or
as the result of charge recombination between Phe⁻ and
 P_{680}^{+} (Barber 1998). Consequently, P_{680}^{+} may play an
indirect cole, in triggering pho as the result of charge recombination between Phe⁻ and P_{680}^{+} (Barber 1998). Consequently, P_{680}^{+} may play an indirect role in triggering photoinactivation, by preventing the quenching of ${}^{3}P_{\infty}^{*}$, by B P_{680}^{+} (Barber 1998). Consequently, P_{680}^{+} may play an indirect role in triggering photoinactivation, by preventing the quenching of ${}^{3}P_{680}^{*}$ by β -carotene while ${}^{3}P_{2}^{*}$ could play a more direct indirect role in triggering photoinactivation, by
preventing the quenching of ${}^{3}P_{680}^{*}$ by β -carotene while
 ${}^{3}P_{680}^{*}$ could play a more direct role, through formation of
 ${}^{1}O_{2}$ (Hideg *et al.* 1994; Oha ${}^{3}P_{680}^{*}$ could play a more direct role, through formation of ${}^{1}O_{2}$ (Hideg *et al.* 1994; Ohad *et al.* 1994). It has been suggested, although not demonstrated, that structural ${}^1\text{O}_2$ (Hideg *et al.* 1994; Ohad *et al.* 1994). It has been suggested, although not demonstrated, that structural characteristics of the PS II reaction centre may exclude O. from the region around P680 (Anderson *e* O_2 from the region around P680 (Anderson et al. 1998), from the region around the region around P680 (Anderson *et al.* 1998), the same way that it apparently is excluded from the characteristics of the PS II reaction centre may exclude O_2 from the region around P680 (Anderson *et al.* 1998), in the same way that it apparently is excluded from the region of the light-harvesting system closest to O_2 from the region around P680 (Anderson *et al.* 1998), in the same way that it apparently is excluded from the region of the light-harvesting system closest to the core complex (Siefermann-Harms $\&$ Angerbofer 1998) in the same way that it apparently is excluded from the
region of the light-harvesting system closest to the core
complex (Siefermann-Harms & Angerhofer 1998). Given
the potential lack of protection from B-carotene, this m region of the light-harvesting system closest to the core complex (Siefermann-Harms & Angerhofer 1998). Given the potential lack of protection from β-carotene, this may complex (Siefermann-Harms & Angerhofer 1998). Given
the potential lack of protection from β -carotene, this may
represent the main mechanism protecting against the
formation of O^* at the reaction centre the potential lack of protection from β -ca
represent the main mechanism protec
formation of ${}^{1}O_{2}^{*}$ at the reaction centre.
Double reduction of the plastoquino

formation of ${}^{1}O_{2}^{*}$ at the reaction centre.
Double reduction of the plastoquinone Q_{A} has also formation of ${}^1O_2^*$ at the reaction centre.
Double reduction of the plastoquinone Q_A has also
been proposed as a potential trigger for photoinactivation
(Van Wijk et al. 1992; Vass et al. 1992). The idea is that Double reduction of the plastoquinone Q_A has also
been proposed as a potential trigger for photoinactivation
(Van Wijk *et al.* 1992; Vass *et al.* 1992). The idea is that
double reduction of Q_A results in formation o been proposed as a potential trigger for photoinactivation
(Van Wijk *et al.* 1992; Vass *et al.* 1992). The idea is that
double reduction of Q_A results in formation of hydroplasto-
quinone (O_H) which is released from (Van Wijk *et al.* 1992; Vass *et al.* 1992). The idea is that double reduction of Q_A results in formation of hydroplasto-quinone ($Q_A H_2$), which is released from its binding site. \bigcup This loss of Q_A could lead to a substantial increase in the quinone $(Q_A H_2)$, which is released from its binding site.
This loss of Q_A could lead to a substantial increase in the
yield of ${}^3P_{680}^*$ through charge recombination. The possibi-
lity that double reduction of Ω This loss of Q_A could lead to a substantial increase in the
yield of ${}^{3}P_{680}^{*}$ through charge recombination. The possibi-
lity that double reduction of Q_A could be the trigger for
photoinactivation, has been str yield of ${}^{3}P_{680}^{*}$ through charge recombination. The possibility that double reduction of Q_A could be the trigger for photoinactivation has been strongly contested on the grounds that target theory reveals photoin Let us that double reduction of Q_A could be the trigger for photoinactivation has been strongly contested on the grounds that target theory reveals photoinactivation to be 1998). a single-photon event (Sinclair *et al.* 1996; Anderson *et al.* 1998).
Photoinactivation of PS II has been shown to follow the

reciprocity law in isolated thylakoids of *Spinacea oleracia*, Photoinactivation of PS II has been shown to follow the
reciprocity law in isolated thylakoids of *Spinacea oleracia*,
in cells of *Anacystis nidulans* (Jones & Kok 1966) and
Synechocystis 6803 (Nagy *et al.* 1995) and i *Synechocystics* 1 aw in isolated thylakoids of *Spinacea oleracia*, ship cells of *Anacystis nidulans* (Jones & Kok 1966) and compress the *Synechocystis* 6803 (Nagy *et al.* 1995), and in leaf discs from the appropriate in cells of *Anacystis nidulans* (Jones & Kok 1966) and
Synechocystis 6803 (Nagy *et al.* 1995), and in leaf discs from
a number of higher plant species (Park *et al.* 1995;
Anderson *et al.* 1998) Consequently photoinac Synechocystis 6803 (Nagy *et al.* 1995), and in leaf discs from
a number of higher plant species (Park *et al.* 1995;
Anderson *et al.* 1998). Consequently, photoinactivation, *in*
i^{ing} appears to be proportional to th a number of higher plant species (Park *et al.* 1995; Anderson *et al.* 1998). Consequently, photoinactivation, *in vivo*, appears to be proportional to the number of photons *Phil. Trans. R. Soc. Lond.* B (2000)
Phil. Trans. R. Soc. Lond. B (2000)

absorbed. For example, 5 h exposure to 500μ mol m⁻²s⁻ absorbed. For example, 5 h exposure to 500 μ mol m⁻²s⁻¹ would be expected to result in the same number of PS II would be expected to result in the same number of PS II units becoming photoinactivated as 1h exposure to would be expected to result in the same number of PS II
units becoming photoinactivated as 1h exposure to
2500 μ mol m⁻²s⁻¹. Anderson *et al.* (1998) have suggested
that this 'light-dose' response of photoinactivati units becoming photoinactivated as 1h exposure to 2500μ mol m⁻²s⁻¹. Anderson *et al.* (1998) have suggested that this 'light-dose' response of photoinactivation implies the existence of a single trigger for photoina 2500 μ mol m⁻²s⁻¹. Anderson *et al.* (1998) have suggested that this 'light-dose' response of photoinactivation implies the existence of a single trigger for photoinactivation.
Clearly the vield of this putative tri that this 'light-dose' response of photoinactivation implies
the existence of a single trigger for photoinactivation.
Clearly, the yield of this putative trigger (the probability
of an absorbed photon inducing formation of the existence of a single trigger for photoinactivation.
Clearly, the yield of this putative trigger (the probability
of an absorbed photon inducing formation of the trigger)
must be largely independent of incident photosy Clearly, the yield of this putative trigger (the probability
of an absorbed photon inducing formation of the trigger)
must be largely independent of incident photosyntheti-
cally active photon flux density (PPED) of an absorbed photon inducing formation of the trigger) must be largely independent of incident photosynthetiust be largely independent of incident photosynthetily
active photon flux density (PPFD).
There are two processes operating at PS II that are
ry likely to play important roles in the regulation of

formation is likely to be highest (Siefermann-Harms & ranging from darkness to full sunlight, the combination of Angerhofer 1998).
As noted above, the oxidizing potential of P_{680}^+ is high $\frac{P_{680}^+}{P_{680}^-}$ (all cally active photon flux density (PPFD).
There are two processes operating at PS II that are
very likely to play important roles in the regulation of
photoinactivation: changes in the canacity for stable There are two processes operating at PS II that are
very likely to play important roles in the regulation of
photoinactivation: changes in the capacity for stable
charge separation (photochemistry) and the effective rate very likely to play important roles in the regulation of
photoinactivation: changes in the capacity for stable
charge separation (photochemistry) and the effective rate
constant for one or more non-radiative processes that photoinactivation: changes in the capacity for stable
charge separation (photochemistry) and the effective rate
constant for one or more non-radiative processes that
compete with photochemistry (downregulation) Changes charge separation (photochemistry) and the effective rate
constant for one or more non-radiative processes that
compete with photochemistry (downregulation). Changes
in the canacity for photochemistry and downregulation at constant for one or more non-radiative processes that
compete with photochemistry (downregulation). Changes
in the capacity for photochemistry and downregulation at
PS II can be monitored through measurement of chlorocompete with photochemistry (downregulation). Changes
in the capacity for photochemistry and downregulation at
PS II can be monitored through measurement of chloro-
phyll fluorescence (reviewed by Krause & Weis 1991; Dau in the capacity for photochemistry and downregulation at
PS II can be monitored through measurement of chloro-
phyll fluorescence (reviewed by Krause & Weis 1991; Dau
1994). Under *in nine* conditions, at incident light l phyll fluorescence (reviewed by Krause & Weis 1991; Dau
1994). Under *in vivo* conditions, at incident light levels phyll fluorescence (reviewed by Krause & Weis 1991; Dau
1994). Under *in vivo* conditions, at incident light levels
ranging from darkness to full sunlight, the combination of
photochemistry and downregulation normally resu 1994). Under *in vivo* conditions, at incident light levels ranging from darkness to full sunlight, the combination of photochemistry and downregulation normally result in a 'quenching' of chlorophyll fluorescence to a st ranging from darkness to full sunlight, the combination of
photochemistry and downregulation normally result in a
"quenching" of chlorophyll fluorescence to a steady-state
vield within a narrow range of hetween ca , 2 and photochemistry and downregulation normally result in a 'quenching' of chlorophyll fluorescence to a steady-state yield within a narrow range of between *ca*. 2 and 4% (Havaux *et al.* 1991; Genty *et al.* 1992; Laisk *et a* ²quenching' of chlorophyll fluorescence to a steady-state yield within a narrow range of between *ca.* 2 and 4% (Havaux *et al.* 1991; Genty *et al.* 1992; Laisk *et al.* 1997). Down regulation has been shown to protect Downregulation has been shown to protect against photo-Downregulation has been shown to protect against photo-
inhibition (Krause & Behrend 1986; Oxborough & Downregulation has been shown to protect against photo-
inhibition (Krause & Behrend 1986; Oxborough &
Horton 1988), presumably by decreasing the yield of the
trigger(s) of photoinactivation inhibition (Krause & Behren
Horton 1988), presumably by de
trigger(s) of photoinactivation.
Charge separation at PS II proton 1988), presumably by decreasing the yield of the
gger(s) of photoinactivation.
Charge separation at PS II is stabilized through the
unsfer of an electron from Phe⁻ to the first stable

trigger(s) of photoinactivation.
Charge separation at PS II is stabilized through the
transfer of an electron from Phe⁻ to the first stable
acceptor Ω . Eurther stable charge separation at PS II Charge separation at PS II is stabilized through the
transfer of an electron from Phe⁻ to the first stable
acceptor, Q_A . Further stable charge separation at PS II
can only occur when P680⁺ and Q^- have returned to transfer of an electron from Phe⁻ to the first stable
acceptor, Q_A . Further stable charge separation at PS II
can only occur when P680⁺ and Q_A^- have returned to the
ground state. Because oxidation of Q_A^- is roug acceptor, Q_A . Further stable charge separation at PS II can only occur when P680⁺ and Q_A^- have returned to the paration at PS II
we returned to the
 $\frac{1}{A}$ is roughly four
eduction of P⁺ can only occur when P680⁺ and Q_A^- have returned to the
ground state. Because oxidation of Q_A^- is roughly four
orders of magnitude slower than reduction of P_{680}^+
(Robinson & Crofts 1983: Meyer *et al.* 1989: Cro ground state. Because oxidation of Q_A^- is roughly four orders of magnitude slower than reduction of P_{680}^+ (Robinson & Crofts 1983; Meyer *et al.* 1989; Crofts *et al.* 1993: Day 1994). PS II centres are described a orders of magnitude slower than reduction of P_{680}^+
(Robinson & Crofts 1983; Meyer *et al.* 1989; Crofts *et al.* 1993; Dau 1994), PS II centres are described as being (Robinson & Crofts 1983; Meyer *et al.* 1989; Crofts *et al.* 1993; Dau 1994), PS II centres are described as being 'open' (capable of stable charge separation) when Q_A is in the ground state and as 'closed' (not capabl 1993; Dau 1994), PS II centres are described as being

'open' (capable of stable charge separation) when Q_A is

in the ground state and as 'closed' (not capable of stable

charge separation) when Q_B is carrying a sing 'open' (capable of stable charge separation) when Q_A is
in the ground state and as 'closed' (not capable of stable
charge separation) when Q_A is carrying a single negative
charge If all PS II centres were isolated fro in the ground state and as 'closed' (not capable of stable
charge separation) when Q_A is carrying a single negative
charge. If all PS II centres were isolated from each other, charge separation) when Q_A is carrying a single negative
charge. If all PS II centres were isolated from each other,
the fluorescence yield above F_0 or F'_0 would be directly
proportional to the fraction of closed c charge. If all PS II centres were isolated from each other,
the fluorescence yield above F_0 or F'_0 would be directly
proportional to the fraction of closed centres $(1 - [Q_A])$.
In reality connectivity among centres res the fluorescence yield above F_0 or F'_0 would be directly
proportional to the fraction of closed centres $(1 - [Q_A])$.
In reality, connectivity among centres results in a curvi-
linear relationship between $1 - [Q_1]$ and proportional to the fraction of closed centres $(1 - [Q_A])$.
In reality, connectivity among centres results in a curvilinear relationship between $I - [Q_A]$ and the fluorescence ity among centres results in a curvi-

etween $1 - [Q_A]$ and the fluorescence
 $\frac{1}{2}$ (Joliot & Joliot 1964). The effect of

orescence parameters is discussed in linear relationship between $1 - [Q_A]$ and the fluorescence
yield above F_o or F_o' (Joliot & Joliot 1964). The effect of
connectivity on fluorescence parameters is discussed in
more detail below $(S.2(b))$ yield above F_0 or F'_0 (Joliot
connectivity on fluorescence
more detail below (§ 2(b)).
Evidence from a large m connectivity on fluorescence parameters is discussed in
more detail below $(\S 2(b))$.
Evidence from a large number of empirical observa-

more detail below ($\S 2(b)$).

Evidence from a large number of empirical observa-

tions suggest that downregulation is dominated by Stern-

Volmer quenching (Lavergne & Trissl 1995). Within the

Stern-Volmer model fluores tions suggest that downregulation is dominated by Stern-Volmer quenching (Lavergne & Trissl 1995). Within the
Stern–Volmer model, fluorescence and quencher concen-
tration are linked through the Stern–Volmer equation Volmer quenching (Lavergne & Trissl 1995). Within the Stern–Volmer model, fluorescence and quencher concentration are linked through the Stern–Volmer equation, which simply states that the reciprocal of fluorescence Stern–Volmer model, fluorescence and quencher concentration are linked through the Stern–Volmer equation, which simply states that the reciprocal of fluorescence vield is proportional to quencher concentration. In tration are linked through the Stern–Volmer equation,
which simply states that the reciprocal of fluorescence
yield is proportional to quencher concentration. In
reality there is little evidence that fluorescence yield is which simply states that the reciprocal of fluorescence yield is proportional to quencher concentration. In reality, there is little evidence that fluorescence yield is actually modulated by quencher concentration and it should be noted that a change in the effective rate constant for a quencher within the pigment bed would actually modulated by quencher concentration and it should be noted that a change in the effective rate constant for a quencher within the pigment bed would have exactly the same effect as a change in quencher should be noted that a change in the effective rate concentration. we exactly the same effect as a change in quencher
ncentration.
The widely used fluorescence parameter, $(F_m/F_m') - 1$, is
rived from the Stern-Volmer equation and can be used

concentration.
The widely used fluorescence parameter, $(F_m/F_m') - 1$, is
derived from the Stern–Volmer equation and can be used

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Figure 1. A schematic showing the different ways in which de-excitation can occur at PS II. (*a*) represents the equilibrated Figure 1. A schematic showing the different ways in which de-excitation can occur at PS II. (*a*) represents the equilibrated excited state within the pigment bed. k_c is the apparent rate constant for charge separation Figure 1. A schematic showing the different ways in which de-excitation can occur at PS II. (*a*) represents the equilibrated excited state within the pigment bed. k_c is the apparent rate constant for charge separation excited state within the pigment bed. k_C is the apparent rate constant for charge separation that leads to formation of (b) the radical pair. k_R is the rate constant for charge recombination, leading to re-formation radical pair. k_R is the rate constant for charge recombination, leading to re-formation of the equilibrated excited state, (*a*).
 k_S is the rate constant for charge stabilization, which is the sum of electron transfer k_S is the rate constant for charge stabilization, which is the sum of electron transfer to Q_A and charge recombination leading to formation of ${}^3P_{680}^*$ or ${}^1P_{680}$. k_F is the rate constant for fluorescence e formation of ${}^{3}P_{680}^{*}$ or ${}^{1}P_{680}$. k_{F} is the rate constant for fluorescence emission. k_{D} is the rate const
occurs in the dark-adapted state. k_{SV} is the rate constant for non-radiative decay that resu

to follow changes in apparent quencher concentration
(Bilger & Biörkman 1991) Dominance of downregulation to follow changes in apparent quencher concentration
(Bilger & Björkman 1991). Dominance of downregulation
by Stern Volmer quenching provides a plausible explanato follow changes in apparent quencher concentration
(Bilger & Björkman 1991). Dominance of downregulation
by Stern–Volmer quenching provides a plausible explana-
tion for the linearity that has often been observed (Bilger & Björkman 1991). Dominance of downregulation
by Stern-Volmer quenching provides a plausible explana-
tion for the linearity that has often been observed
between the fluorescence parameter F'/F' (which is by Stern–Volmer quenching provides a plausible explana-
tion for the linearity that has often been observed
between the fluorescence parameter F'_{q}/F'_{m} (which is
often written as $\Delta F/F'$ in the literature) and the between the fluorescence parameter F'_q/F'_m (which is often written as $\Delta F/F'_m$ in the literature) and the between the fluorescence parameter F'_{q}/F'_{m} (which is
often written as $\Delta F/F'_{m}$ in the literature) and the
quantum yield of CO₂ assimilation (ϕ_{CO_2}). This relation-
ship was first described by Genty *et al* (1 often written as $\Delta F/F_m'$ in the literature) and the quantum yield of CO_2 assimilation (ϕ_{CO_2}). This relationship was first described by Genty *et al.* (1989) and has been verified since by data from a number of oth quantum yield of CO_2 assimilation (ϕ_{CO_2}). This relationship was first described by Genty *et al.* (1989) and has been verified since by data from a number of other groups (e.g. Di. Marco. *et al.* 1990; Krall & Edw ship was first described by Genty *et al.* (1989) and has
been verified since by data from a number of other groups
(e.g. Di Marco *et al.* 1990; Krall & Edwards 1990;
Edwards & Baker 1993); indeed this relationship provid (e.g. Di Marco et al. 1990; Krall & Edwards 1990; (e.g. Di Marco *et al.* 1990; Krall & Edwards 1990; Edwards & Baker 1993); indeed this relationship provides perhaps the strongest empirical evidence that down-regulation is dominated by Stern-Volmer quenching regulation is dominated by Stern-Volmer quenching (Lavergne & Trissl 1995). perhaps the strongest em
regulation is dominated
(Lavergne & Trissl 1995).
 F'/F' provides an estima

gulation is dominated by Stern–Volmer quenching

avergne & Trissl 1995).
 F_q'/F_m' provides an estimate of the operating efficiency of

otochemistry at PS II and is actually the product of two photochemistry at PS II and is actually the product of two centres increases at moderate to high light levels, the other fluorescence parameters, F_v/F_m and F_a/F_v . F_v/F_m lower rate of charge separation at closed centres $F'_{\rm q}/F'_{\rm m}$ provides an estimate of the operating efficiency of photochemistry at PS II and is actually the product of two other fluorescence parameters, $F'_{\rm y}/F'_{\rm m}$ and $F'_{\rm q}/F'_{\rm y}$. $F'_{\rm y}/F'_{\rm p}$ provid photochemistry at PS II and is actually the product of two

other fluorescence parameters, F_v'/F_m' and F_q'/F_v' . F_v'/F_m'

provides an estimate of what the maximum efficiency of

photochemistry at PS II would be in the l other fluorescence parameters, F_v/F_m and F_q/F_v . F_v/F_m
provides an estimate of what the maximum efficiency of
photochemistry at PS II would be, in the light-adapted
state if all centres were open ([O, 1 – 1) F'/F' is a provides an estimate of what the maximum efficiency of
photochemistry at PS II would be, in the light-adapted
state, if all centres were open $([\mathbf{Q}_A] = 1)$. F'_q/F'_v is a factor
that relates the maximum and operating eff state, if all centres were open ($[Q_A] = 1$). F'_q/F'_v is a factor
that relates the maximum and operating efficiencies of
photochemistry at PS II and is mathematically the same
as the widely used 'coefficient of photochemi photochemistry at PS II would be, in the light-adapted
state, if all centres were open ($[Q_A] = 1$). F'_q/F'_v is a factor
that relates the maximum and operating efficiencies of that relates the maximum and operating efficiencies of
photochemistry at PS II and is mathematically the same
as the widely used 'coefficient of photochemical quenching'
 (a_n) . Use of the term a_n has been avoid (q_P) . Use of the term q_P has been avoided here because, in btochemistry at PS II and is mathematically the same
the widely used 'coefficient of photochemical quenching'
). Use of the term q_P has been avoided here because, in
context of a Stern Volmer model for downregulati as the widely used 'coefficient of photochemical quenching' (q_P). Use of the term q_P has been avoided here because, in the context of a Stern–Volmer model for downregulation, (q_P). Use of the term q_P has been avoided here because, in
the context of a Stern–Volmer model for downregulation,
 F'_q/F'_v is a factor (rather than a coefficient). Also, q_P has
heen widely used either as a provy fo the context of a Stern-Volmer model for downregulation,
 F_q/F_v' is a factor (rather than a coefficient). Also, q_p has
been widely used either as a proxy for $[Q_A]$ or as a semi-
quantitative indicator of $[Q_A]$ neither F'_q/F'_v is a factor (rather than a coefficient). Also, q_P has
been widely used either as a proxy for $[Q_A]$ or as a semi-
quantitative indicator of $[Q_A]$, neither of which is reason-
able within a model where downregu quantitative indicator of $[Q_A]$, neither of which is reasonable within a model where downregulation is dominated
by Stern-Volmer quenching and there is also a high level
of connectivity among PS II centres (see δ ?) able within a model where downregulation is
by Stern-Volmer quenching and there is also a
of connectivity among PS II centres (see $\S 2$).
A number of different models (reviewed by Stern-Volmer quenching and there is also a high level
connectivity among PS II centres (see $\S 2$).
A number of different models (reviewed by Dau 1994)
we been proposed to relate the de-excitation processes

% of connectivity among PS II centres (see § 2).
A number of different models (reviewed by Dau 1994) have been proposed to relate the de-excitation processes A number of different models (reviewed by Dau 1994)
have been proposed to relate the de-excitation processes
that operate within PS II (Butler & Kitajima 1975; Butler
1978; Schatz et al. 1988; Laible et al. 1994; Dau & Sa have been proposed to relate the de-excitation processes
that operate within PS II (Butler & Kitajima 1975; Butler
1978; Schatz *et al.* 1988; Laible *et al.* 1994; Dau & Sauer
1996). An increasing amount of evidence, most that operate within PS II (Butler & Kitajima 1975; Butler us
1978; Schatz *et al.* 1988; Laible *et al.* 1994; Dau & Sauer mostly from 1996). An increasing amount of evidence, mostly from ra 1978; Schatz *et al.* 1988; Laible *et al.* 1994; Dau & Sauer
1996). An increasing amount of evidence, mostly from
picosecond fluorescence studies (Schatz *et al.* 1988;
McCauley *et al.* 1989; Roelofs *et al.* 1992; Dau & 1996). An increasing amount of evidence, mostly from picosecond fluorescence studies (Schatz *et al.* 1988; McCauley *et al.* 1989; Roelofs *et al.* 1992; Dau & Sauer 1996) strongly supports the reversible radical pair (RR picosecond fluorescence studies (Schatz *et al.* 1988;
McCauley *et al.* 1989; Roelofs *et al.* 1992; Dau & Sauer
1996), strongly supports the reversible radical pair (RRP)
equilibrium model originally proposed by Schatz McCauley *et al.* 1989; Roelofs *et al.* 1992; Dau & Sauer 1996), strongly supports the reversible radical pair (RRP) equilibrium model, originally proposed by Schatz *et al.* 1996), strongly supports the reversible radical pair (RRP) is equilibrium model, originally proposed by Schatz *et al.*
(1988). The 'equilibrium' part of the RRP equilibrium k_S model refers to a rapid equilibriation of equilibrium model, originally proposed by Schatz *et al.* (1988). The 'equilibrium' part of the RRP equilibrium model refers to a rapid equilibration of excited states that is established among the chlorophylls within the (1988). The 'equilibrium' part of the RRP equilibrium
model refers to a rapid equilibration of excited states that
is established among the chlorophylls within the PS II is established among the chlorophylls within the PS II
Phil. Trans. R. Soc. Lond. B (2000)

complex, including P680, within a much shorter time-
frame $(c_4, 15 \text{ ps})$, than the mean lifetime of an exciton complex, including P680, within a much shorter time-
frame (*ca*. 15 ps), than the mean lifetime of an exciton
 (5 log) (Schatz *et al.* 1988: Day & Sayer 1996). The complex, including P680, within a much shorter time-
frame $(caa$. 15 ps), than the mean lifetime of an exciton
 $(> \ln s)$ (Schatz *et al.* 1988; Dau & Sauer 1996). The
'RRP' part of the model refers to the possibility that frame (*ca.* 15 ps), than the mean lifetime of an exciton ($>$ 1 ns) (Schatz *et al.* 1988; Dau & Sauer 1996). The 'RRP' part of the model refers to the possibility that ($> 1 \text{ ns}$) (Schatz *et al.* 1988; Dau & Sauer 1996). The 'RRP' part of the model refers to the possibility that charge recombination between P_{680}^+ and Phe⁻ can result in re-formation of the singlet excited state **THERP** part of the model refers to the possibility that charge recombination between P_{680}^+ and Phe⁻ can result in re-formation of the singlet excited state of P680 (${}^{1}P_{680}^*$) and subsequent re-coulibration o charge recombination between P_{680}^{+} and Phe⁻ can result
in re-formation of the singlet excited state of P680 (${}^{1}P_{680}^{*}$)
and subsequent re-equilibration of the excitation among
the chlorophylls within the PS in re-formation of the singlet excited state of P680 (P_{680}^*)
and subsequent re-equilibration of the excitation among
the chlorophylls within the PS II complex.
Data from picosecond fluorescence studies also indicate d subsequent re-equilibration of the excitation among
e chlorophylls within the PS II complex.
Data from picosecond fluorescence studies also indicate
at charge separation between P680 and Phe is strongly

that charge separation between P680 and Phe is strongly inhibited at closed PS II centres (Schatz *et al.* 1988; McCauley *et al.* 1989; Roelofs *et al.* 1992), a view that is inhibited at closed PS II centres (Schatz *et al.* 1988;
McCauley *et al.* 1989; Roelofs *et al.* 1992), a view that is
also supported by the electron paramagnetic resonance
(EPR) and flash spectroscopic measurements of V McCauley *et al.* 1989; Roelofs *et al.* 1992), a view that is
also supported by the electron paramagnetic resonance
(EPR) and flash spectroscopic measurements of Van
Meighem *et al.* (1995). Since the fraction of closed also supported by the electron paramagnetic resonance (EPR) and flash spectroscopic measurements of Van Meighem *et al.* (1995). Since the fraction of closed PS II centres increases at moderate to birsh light levels the (EPR) and flash spectroscopic measurements of Van Meighem et al. (1995) . Since the fraction of closed PS II centres increases at moderate to high light levels, the
lower rate of charge separation at closed centres must be
taken into account when considering likely mechanisms
for the triggering of photoinactivation in the context for the triggering separation at closed centres must be taken into account when considering likely mechanisms
for the triggering of photoinactivation, in the context of a
light-dose response for this process taken into account when considerin
for the triggering of photoinactivatio
light-dose response for this process.

2. MATERIAL AND METHODS (a) *Rate constants*

been widely used either as a proxy for $[Q_A]$ or as a semi-

able within a model where downregulation is dominated

able within a model where downregulation is dominated describe their 'bipartite' model, which is homologo (a) **Rate constants**
The method of analysis used in this study is based on prob-(a) **Rate constants**
The method of analysis used in this study is based on prob-
abilities, expressed in terms of rate constants, for each of the
possible de sucitation pethusus. As such it is exertially the The method of analysis used in this study is based on probabilities, expressed in terms of rate constants, for each of the possible de-excitation pathways. As such, it is essentially the possible and a proposal bundle Rut abilities, expressed in terms of rate constants, for each of the
possible de-excitation pathways. As such, it is essentially the
same as the approach used by Butler & Kitajima (1975) to
describe their 'binarity' model, whi possible de-excitation pathways. As such, it is essentially the
same as the approach used by Butler & Kitajima (1975) to
describe their 'bipartite' model, which is homologous to the
P.P.B. model (Dou 1994) Deviations of th describe their 'bipartite' model, which is homologous to the RRP model (Dau 1994). Derivations of the equations used are presented in Appendix B. A schematic of the model used is shown in figure 1. presented in Appendix B. A schematic of the model used is

The data presented here are derived from calculations that shown in figure 1.
The data presented here are derived from calculations that
use rate constants calculated for α -centres within isolated
mambranes by Poolefs et al. (1002). The in vira values of these The data presented here are derived from calculations that
use rate constants calculated for α -centres within isolated
membranes by Roelofs *et al.* (1992). The *in vivo* values of these membranes by Roelofs *et al.* (1992). The *in vivo* values of these rate constants may be somewhat different. However, they would membranes by Roelofs *et al.* (1992). The *m vivo* values of these
rate constants may be somewhat different. However, they would
have to be markedly different for any of the conclusions reached
in this paper to be involida rate constants may be somewhat different. However, they would
have to be markedly different for any of the conclusions reached
in this paper to be invalidated. A complete set of the values used
is given in table l in this paper to be invalidated. A complete set of the values used
is given in table 1. this paper to be invalidated. A complete set of the values used
given in table 1.
An arbitrary value of $1000 \mu s^{-1}$ was selected for the value of
 Λs noted in $S1$, the 'ennanced'. Starr, Valmar, quanding

is given in table 1.

An arbitrary value of $1000 \mu s^{-1}$ was selected for the value of k_{SV} . As noted in §1, the 'apparent' Stern–Volmer quenching sould actually result from a change in the rate constant, rather k_{SV} . As noted in §1, the 'apparent' Stern-Volmer quenching could actually result from a change in the rate constant, rather than quencher concentration. Consequently, it would have been could actually result from a change in the rate constant, rather

Table 1. *Values of the rate constants used in this study*

Table 1. *Values of the rate constants used in this study*
(The values of k_C , k_S , k_R and k_D are those calculated for α -centres by Roelofs *et al.* (1992). The values of k_F and k_D were selected to give a flu α -centres by Roelofs *et al.* (1992). The values of k_F and were selected to give a fluorescence yield of 2% at F_o and observed value of F_v'/F_m . The value of k_{SV} is arbitrary.)

equally valid to assign a constant arbitrary value to [SV] and vary the value of k_{SV} . between the value of k_{SV} .

wary the value of k_{SV} .

With each data set, the values of k_{F} and k_{D} were selected,

through iteration to give a fluorescape viald of 2% at the deals

vary the value of k_{SV} .
With each data set, the values of k_{F} and k_{D} were selected,
through iteration, to give a fluorescence yield of 2% at the dark-
adopted E and the observed value of E (E. The value With each data set, the values of k_F and k_D were selected,
through iteration, to give a fluorescence yield of 2% at the dark-
adapted F_0 and the observed value of F_v/F_m . The value of F'_0 ,
which is not wised for through iteration, to give a fluorescence yield of 2% at the dark-
adapted F_0 and the observed value of F_v/F_m . The value of F'_0 ,
which is required for calculation of F'_v/F'_m and F'_q/F'_v , was calcu-
lated using the adapted F_0 and the observed value of F_v/F_m . The value of F'_0 ,
which is required for calculation of F'_v/F'_m and F'_q/F'_v , was calculated using the method of Oxborough & Baker (1997), using the lated using the method of Oxborough & Baker (1997), using the values of F_0 and F_m measured at the first saturating pulse. At lated using the method of Oxborough & Baker (1997), using the
values of F_0 and F_m measured at the first saturating pulse. At
each data point, values of [SV] and [Q_A] were selected,
through iteration to match with t values of F_0 and F_m measured at the first saturating pulse. A
each data point, values of [SV] and [Q_A] were selected
through iteration, to match with the calculated values of F_v'/F_v through iteration, to match with the calculated values of F_v/F'_m and F'_q/F'_v .

(b) *Connectivity among PS II centres*

It has long been appreciated that the level of connectivity among PS II centres affects the relationship between $1-[Q_A]$ It has long been appreciated that the level of connectivity
among PS II centres affects the relationship between $1 - [Q_A]$
(the proportion of closed PS II centres) and ϕ_F (Joliot & Joliot
1064: Patlan & Kitaiima 1075: L among PS II centres affects the relationship between $1 - [Q_A]$
(the proportion of closed PS II centres) and ϕ_F (Joliot & Joliot
1964; Butler & Kitajima 1975; Lavergne & Trissl 1995). In the
featured probability where ap (the proportion of closed PS II centres) and ϕ_F (Joliot & Joliot 1964; Butler & Kitajima 1975; Lavergne & Trissl 1995). In the 'separate package' model, where energy transfer among PS II 1964; Butler & Kitajima 1975; Lavergne & Trissl 1995). In the

'separate package' model, where energy transfer among PS II

units is not possible, the relationship between $1 - [Q_A]$ and ϕ_F is

linear With increasing con Separate package' model, where energy transfer among PS II
units is not possible, the relationship between $1 - [Q_A]$ and ϕ_F is
linear. With increasing connectivity, the relationship becomes
increasingly complinear with **i**ncreasing connectivity, the relationship becomes increasingly curvilinear with ϕ_F being lower than $1 - [Q_A]$ at linear. With increasing connectivity, the relationship becomes
increasingly curvilinear with ϕ_F being lower than $1 - [Q_A]$ at
intermediate values for $[Q_A]$ (Joliot & Joliot 1964). If energy
transfor among PS II with is increasingly curvilinear with ϕ_F being lower than $l - [Q_A]$ at intermediate values for $[Q_A]$ (Joliot & Joliot 1964). If energy transfer among PS II units is unrestricted (a 'lake' model) Q_A behaves as a Stern-Volmer

transfer among PS II units is unrestricted (a 'lake' model) Q_A
behaves as a Stern-Volmer quencher (Shinkarev & Govindjee
1993). Although the equations used here only consider the lake
model for appear transfer abonsing behaves as a Stern-Volmer quencher (Shinkarev & Govindjee
1993). Although the equations used here only consider the lake
model for energy transfer, changing the level of connectivity
within the model would not effect any o 1993). Although the equations used here only consider the lake
model for energy transfer, changing the level of connectivity
within the model would not affect any of the relationships
sumined in this study. model for energy transf
within the model woule
examined in this study.

(c) *Experimental*

(i) *Growing of plants*

Chamber-grown plants of maize (*Zea maize* cv. LG 20.80), bean (*i*) *Growing of plants*

Chamber-grown plants of maize (*Zea maize* cv. LG 20.80), bean

(*Phaseolus vulgaris*) and commelina (*Commelina communis*) were used

fact the sumpriments described have Conscience and itises fo $(Phaseolus vulgaris)$ and commelina $(Commelina communis)$ were used for the experiments described here. Growing conditions for

all three species were the same as described for maize in Oxborough & Baker (1997).

(ii) *Measurement of chlorophyll fluorescence*

Measurement of chlorophyll fluorescence
All fluorescence measurements were made using a Hansatech
489 fluoremeter (Horsetzek Instruments Ltd, Norfoll, LIK) (ii) *Measurement of chlorophyll fluorescence*
All fluorescence measurements were made using a Hansatech
FMS2 fluorometer (Hansatech Instruments Ltd, Norfolk, UK).
Deta ware agguined wing Hansatech ElyonChart agfunna FMS2 fluorometer (Hansatech Instruments Ltd, Norfolk, UK).
Data were acquired using Hansatech FluorChart software FMS2 fluorometer (Hansatech Instruments Ltd, Norfolk, UK).
Data were acquired using Hansatech FluorChart software
running under Windows 98[©] on a notebook computer. Plants Data were acquired using Hansatech FluorChart software
running under Windows 98° on a notebook computer. Plants
were dark adapted in a growth chamber for at least 1h at 22 °C
hefone magazurements were mode at the sam were dark adapted in a growth chamber for at least 1 h at 22 °C
before measurements were made at the same temperature. After were dark adapted in a growth chamber for at least 1 h at 22 °C
before measurements were made at the same temperature. After
an initial measurement of F_v/F_m , saturation pulses of 900 ms
duration were applied at 5 min. in before measurements were made at the same temperature. After
an initial measurement of F_v/F_m , saturation pulses of 900 ms
duration were applied at 5 min intervals throughout the light
curve Additional measurements of F/E an initial measurement of F_v/F_m , saturation pulses of 900 ms
duration were applied at 5 min intervals throughout the light
curve. Additional measurements of F_v/F_m were made at points
during the light gunus of the light duration were applied at 5 min intervals throughout the light
curve. Additional measurements of F_v/F_m were made at points
during the light curve, after 15 min dark adaptation. The
interval halo we have a fake EMS were us during the light curve, after 15 min dark adaptation. The internal halogen lamp of the FMS was used as the light source in all cases. Measurements were always made from the lowest internal halogen lamp of the FMS was used as the light source light intensity to the highest. The light intensity was increased in all cases. Measurements were always made from the lowest
light intensity to the highest. The light intensity was increased
when the fluorescence yield at $F'_{\rm m}$ was within 0.8% of the
provious value and within 1.9% light intensity to the highest. The light intensity was increased
when the fluorescence yield at F'_{m} was within 0.8% of the
previous value and within 1.2% at *F*, with a minimum of three
makes at seath light intens previous value and within 1.2% at F , with a minimum of three pulses at each light intensity. A sample trace is shown in previous value and within 1.2% at *F*, with a minimum of three
pulses at each light intensity. A sample trace is shown in
figure 2. Although calculated values of F'_0 were used for all of
the data presented F'_c was als pulses at each light intensity. A sample trace is shown in
figure 2. Although calculated values of F'_0 were used for all of
the data presented, F'_0 was also measured during 5 s far-red light
treatment 45 s often select the data presented, F'_0 was also measured during 5 s far-red light treatment, 45 s after each saturating pulse.

3. RESULTS

3. RESULTS
All of the data presented are derived from a single light
rye of commelina (the full fluorescence curve is shown S. RESULTS

All of the data presented are derived from a single light

curve of commelina (the full fluorescence curve is shown

in figure 2). Identical treatments with more than 20 other All of the data presented are derived from a single light
curve of commelina (the full fluorescence curve is shown
in figure 2). Identical treatments with more than 20 other
plants (maize, bean, and, commelina), produced v curve of commelina (the full fluorescence curve is shown
in figure 2). Identical treatments with more than 20 other
plants (maize, bean and commelina) produced very in figure 2). Identical treatments with more than 20 other
plants (maize, bean and commelina) produced very
similar data sets. For the data presented, F'_0 was always
calculated using the values of F_1 and F_2 from t plants (maize, bean and commelina) produced very
similar data sets. For the data presented, F'_0 was always
calculated using the values of F_0 and F_m from the initial
 F_v/F_m pulse and the value of F'_m at the point calculated using the values of F_o and F_m from the initial F_v/F_m pulse and the value of F'_m at the point of calculation (Oxborough & Baker 1997). Basing the analysis on values of F' that were calculated using the F_v/F_m pulse and the value of F'_m at the point of calculation (Oxborough & Baker 1997). Basing the analysis on values of F'_o that were calculated using the nearest appro-
priate values of F and F or using measured tion (Oxborough & Baker 1997). Basing the analysis on values of F'_{o} that were calculated using the nearest appropriate values of F_{o} and F_{m} or using measured values of F'_{o} did not produce data that values of F'_0 that were calculated using the nearest appropriate values of F'_0 and F_m or using measured values of F'_0 did not produce data that were significantly different from those presented priate values of F_{o} and
did not produce data
from those presented.

om those presented.

(a) P_{680}^{+} *as a potential trigger of photoinactivation*

Anderson *et al.* (1999) hous resently syggeted that the

(a) P_{680}^{+} *as a potential trigger of photoinactivation*
Anderson *et al.* (1998) have recently suggested that the
cinrecity observed between light dosage and photo-(a) P_{680}^+ as a potential trigger of photoinactivation
Anderson *et al.* (1998) have recently suggested that the
reciprocity observed between light dosage and photo-
inactivation (Jones & Kok 1966; Park *et al.* 1995) Anderson *et al.* (1998) have recently suggested that the reciprocity observed between light dosage and photo-
inactivation (Jones & Kok 1966; Park *et al.* 1995) can be
accommodated within a model in which photoreciprocity observed between light dosage and photo-
inactivation (Jones & Kok 1966; Park *et al.* 1995) can be
accommodated within a model in which photo-
inactivation is triggered by the presence of P^+ . This inactivation (Jones & Kok 1966; Park *et al.* 1995) can be
accommodated within a model in which photo-
inactivation is triggered by the presence of P_{680}^+ . This
possibility is considered bere accommodated within a model in which photo-
inactivation is triggered by the presence of P_{680}^+ . This
possibility is considered here.
Data from a number of fluorescence studies provide a inactivation is triggered by the presence of P_{680}^+ . This

possibility is considered here.
Data from a number of fluorescence studies provide a
range of values for the rate constants of each of the path-
wave within the RRP equilibrium model (e.g. Schatz et al. Data from a number of fluorescence studies provide a
range of values for the rate constants of each of the path-
ways within the RRP equilibrium model (e.g. Schatz *et al.*
1988: Roelofs *et al.* 1992: Day & Sauer 1996), A range of values for the rate constants of each of the path-
ways within the RRP equilibrium model (e.g. Schatz *et al.* 1988; Roelofs *et al.* 1992; Dau & Sauer 1996). As noted in
the introduction all of these studies indi ways within the RRP equilibrium model (e.g. Schatz *et al.* 1988; Roelofs *et al.* 1992; Dau & Sauer 1996). As noted in the introduction, all of these studies indicate that the rate constant for charge separation at open 1988; Roelofs et al. 1992; Dau & Sauer 1996). As noted in in the terminology used here) is much higher than for constant for charge separation at open PS II centres ($k_{C(0)}$)
in the terminology used here) is much higher than for
charge separation at closed centres ($k_{C(c)}$); a result that is
in agreement with the EPR and flash spe in the terminology used here) is much higher than for
charge separation at closed centres $(k_{C(c)})$; a result that is
in agreement with the EPR and flash spectroscopic
measurements of Van Mieghem et al. (1995). In the charge separation at closed centres $(k_{C(c)})$; a result that is
in agreement with the EPR and flash spectroscopic
measurements of Van Mieghem *et al.* (1995). In the
absence of downregulation, the proportion of centres in in agreement with the EPR and flash spectroscopic
measurements of Van Mieghem *et al.* (1995). In the
absence of downregulation, the proportion of centres in the open state $([Q_A])$ would be expected to decrease absence of downregulation, the proportion of centres in
the open state $([Q_A])$ would be expected to decrease
with increasing PPFD. However, the relationship between
incident PPFD and $[Q_A]$ is complicated by the increase the open state $([Q_A])$ would be expected to decrease
with increasing PPFD. However, the relationship between
incident PPFD and $[Q_A]$ is complicated by the increase
in downregulation with incident PPFD incident PPFD and $[Q_A]$ is complicated by the increase in downregulation with incident PPFD. Exercise in a Fig. 1 is complicated by the increase
downregulation with incident PPFD.
In assessing the likelihood that P_{680}^+ is a trigger of
optimactivation it is important to consider the

in downregulation with incident PPFD.
In assessing the likelihood that P_{680}^+ is a trigger of
photoinactivation, it is important to consider the

Figure 2. Fluorescence trace from an attached commelina leaf, from which all of the data in figures 3 and 4 are derived. T
numbers within the frame of the graph show the incident PPFD (µmol m⁻² s⁻¹) at different point ŏ Figure 2. Fluorescence trace from an attached commelina leaf, from which all of the data in figures 3 and 4 are derived. The Figure 2. Fluorescence trace from an attached commelis
numbers within the frame of the graph show the inciden
Growing and measuring conditions are detailed in $\S 2$. Growing and measuring conditions are detailed in §2.
integrated lifetime of this radical, which is given by the

product of yield and lifetime. The integrated lifetime of integrated lifetime of this radical, which is given by the product of yield and lifetime. The integrated lifetime of P_{680}^+ formed through charge separation is dependent on the way in which it is taken back to the gro product of yield and lifetime. The integrated lifetime of P_{680}^+ formed through charge separation is dependent on the way in which it is taken back to the ground state, which in turn is very largely dependent on wheth P_{680}^{+} formed through charge separation is dependent on the way in which it is taken back to the ground state, which, in turn, is very largely dependent on whether the centre is onen or closed the way in which it is to
which, in turn, is very laid
centre is open or closed.
At open centres charge hich, in turn, is very largely dependent on whether the
tree is open or closed.
At open centres, charge separation is most frequently
lowed by charge stabilization (see δ l) which involves

Followed by charge stabilization (see § 1), which involves followed by charge stabilization (see § 1), which involves At open centres, charge separation is most frequently
followed by charge stabilization (see § 1), which involves
the transfer of an electron from Z to P₆₈₀. In the
remaining cases P⁺₊ at open centres is taken back t followed by charge stabilization (see § 1), which involves
the transfer of an electron from Z to P_{680}^+ . In the
remaining cases, P_{680}^+ at open centres is taken back to the
ground state through charge recombinatio the transfer of an electron from Z to P_{680}^+ . In the remaining cases, P_{680}^+ at open centres is taken back to the ground state through charge recombination. Conversely, P_{+}^{+} at closed centres is nearly always remaining cases, P_{680}^{+} at open centres is taken back to the ground state through charge recombination. Conversely, P_{680}^{+} at closed centres is nearly always taken back to the ground state through charge recombi ground state through charge recombination. Conversely, P_{680}^{+} at closed centres is nearly always taken back to the ground state through charge recombination.
The time constant for electron transfer from Z to $P_{680}^{$ P_{680}^{+} at closed centres is nearly always taken back to the

is between 20 and 300 ns, depending on the current The time constant for electron transfer from Z to P_{680}^+
is between 20 and 300 ns, depending on the current
S-state of the oxygen-evolving complex, with a mean
value of ca , 90 ns (Deprez et al. 1983; Meyer et al. 198 is between 20 and 300 ns, depending on the current S-state of the oxygen-evolving complex, with a mean value of *ca*. 90 ns (Deprez *et al.* 1983; Meyer *et al.* 1989).
Assessing the lifetime of P^+ in situations where S-state of the oxygen-evolving complex, with a mean
value of *ca*. 90 ns (Deprez *et al.* 1983; Meyer *et al.* 1989).
Assessing the lifetime of P_{680}^{+} in situations where it is
taken back to the ground state through value of *ca*. 90 ns (Deprez *et al.* 1983; Meyer *et al.* 1989).
Assessing the lifetime of P_{680}^{+} in situations where it is taken back to the ground state through charge recombina-
tion is complicated by the fact th Assessing the lifetime of P_{680}^{+} in situations where it is
taken back to the ground state through charge recombina-
tion is complicated by the fact that charge recombination
can result in formation of ${}^{1}P_{-}^{*}$, taken back to the ground state through charge recombina-
tion is complicated by the fact that charge recombination
can result in formation of ${}^{1}P_{680}^{*}$, ${}^{3}P_{680}^{*}$ or ${}^{1}P_{680}$. The time
constants for format tion is complicated by the fact that charge recombination
can result in formation of ${}^{1}P_{680}^{*}$, ${}^{3}P_{680}^{*}$ or ${}^{1}P_{680}$. The time
constants for formation of ${}^{1}P_{680}^{*}$ through charge recombi-
pation at can result in formation of ${}^{1}P_{680}^{*}$, ${}^{3}P_{680}^{*}$ or ${}^{1}P_{680}^{*}$. The time
constants for formation of ${}^{1}P_{680}^{*}$ through charge recombi-
nation at open and closed centres are given by $1/k_{R(o)}$ and
 $1/k$ constants for formation of ${}^{1}P_{680}^{*}$ through charge recombi-
nation at open and closed centres are given by $1/k_{R(0)}$ and
 $1/k_{R(c)}$, which provide values of 3.3 and 2.9 ns, respect-
ively. Charge recombination giving nation at open and closed centres are given by $1/k_{R(o)}$ and $1/k_{R(o)}$, which provide values of 3.3 and 2.9 ns, respectively. Charge recombination giving rise to formation of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ cannot be disti ively. Charge recombination giving rise to formation of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ cannot be distinguished from charge stabilization, since all three pathways for de-excitation give rise to non-fluorescence states ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ cannot be disting
stabilization, since all three pathw
give rise to non-fluorescence states.
At open centres, the yield of ${}^{3}P^{*}$ bilization, since all three pathways for de-excitation
ve rise to non-fluorescence states.
At open centres, the yield of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through
arge recombination is unlikely to be very significant

give rise to non-fluorescence states.
At open centres, the yield of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through
charge recombination is unlikely to be very significant,
because the yield of competing charge stabilization is At open centres, the yield of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through
charge recombination is unlikely to be very significant,
because the yield of competing charge stabilization is
known to be very high. At closed centr charge recombination is unlikely to be very significant,
because the yield of competing charge stabilization is
known to be very high. At closed centres, formation of
 ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through charge recomb known to be very high. At closed centres, formation of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through charge recombination almost certainly represent the main post-charge separation, non-fluorescence, pathways for de-excitation ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through charge recombination almost
certainly represent the main post-charge separation,
non-fluorescence pathways for de-excitation, since the *Phil. Trans. R. Soc. Lond.* B (2000)

constants and method used to calculate values of $\phi_{S(0)}$ are
given in §2.
only alternative pathways that have been established are **Figure 3.** The relationship between incident PPFD and the vield of charge stabilization at onen PS II centres (ϕ_{max}) Figure 3. The relationship between incident PPFD and the yield of charge stabilization at open PS II centres $(\phi_{S(0)})$, derived from the fluorescence trace in figure 2, over a range Figure 3. The relationship between incident PPFD and the
yield of charge stabilization at open PS II centres $(\phi_{S(0)})$,
derived from the fluorescence trace in figure 2, over a range of
PPEDs between 0 and 2740 umol m⁻²s yield of charge stabilization at open PS II ce
derived from the fluorescence trace in figure
PPFDs between 0 and 2740 μ mol m⁻² s⁻¹. D
constants and method used to calculate value I centres $(\phi_{S(o)})$,
ure 2, over a range of
. Details of the rate
alues of ϕ derived from the fluorescence trace in figure 2, over a range eppFDs between 0 and 2740 μ mol m⁻²s⁻¹. Details of the rate constants and method used to calculate values of $\phi_{S(0)}$ are given in §2.

only alternative pathways that have been established are electron transfer to $Q_A^-(\text{Van Wijk} \text{ et al. } 1992)$; Vass *et al.* 1999) and to evtochrome *h* (Poulson *et al.* 1995) peither only alternative pathways that have been established are electron transfer to Q_A^- (Van Wijk *et al.* 1992; Vass *et al.* 1992) and to cytochrome b_{559} (Poulson *et al.* 1995), neither of which is thought capable of s electron transfer to Q_A^- (Van Wijk *et al.* 1992; Vass *et al.* 1992) and to cytochrome b_{559} (Poulson *et al.* 1995), neither of which is thought capable of supporting significant rates of electron flow (1992) and to cyto of which is though
of electron flow.
The overall tire which is thought capable of supporting significant rates
electron flow.
The overall time constant for formation of ${}^{3}P_{680}^{*}$ and
through charge recombination, plus charge stabiliza-

of electron flow.
The overall time constant for formation of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through charge recombination, plus charge stabiliza-
tion at onen centres is given by $1/k$ which has a value The overall time constant for formation of ${}^{3}P_{680}^{*}$ and ${}^{1}P_{680}$ through charge recombination, plus charge stabilization at open centres, is given by $1/k_{S(0)}$, which has a value of 0.43 ns. At closed centres, ¹P₆₈₀ through charge recombination, plus charge stabilization at open centres, is given by $1/k_{S(o)}$ which has a value of 0.43 ns. At closed centres, the equivalent time constant is given by $1/k_{S(o)}$ which has a value tion at open centres, is given by $1/k_{S(0)}$, which
of 0.43 ns. At closed centres, the equivalent t
is given by $1/k_{S(0)}$, which has a value of 1 ns.
Clearly, the lifetime of P⁺₊, is very mu 0.43 ns. At closed centres, the equivalent time constant
given by $1/k_{S(c)}$, which has a value of 1 ns.
Clearly, the lifetime of P_{680}^+ is very much longer at
ntres that have undergone charge stabilization than at

is given by $1/k_{S(c)}$, which has a value of 1 ns.
Clearly, the lifetime of P_{680}^+ is very much longer at centres that have undergone charge stabilization than at centres where charge recombination has occurred (by a Clearly, the lifetime of P_{680}^+ is very much longer at centres that have undergone charge stabilization than at centres where charge recombination has occurred (by a factor of approximately 30). Consequently, the inte centres that have undergone charge stabilization than at
centres where charge recombination has occurred (by a
factor of approximately 30). Consequently, the integrated
lifetime of P_{max}^{\pm} , at centres, that undergo centres where charge recombination has occurred (by a factor of approximately 30). Consequently, the integrated lifetime of P_{680}^{+} at centres that undergo charge

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would be in the absence of downregulation.

recombination will only be significant if the yield of Figure 4. Data illustrating the effect of downregulation of the relationships between the yield of charge stabilization at open PS Figure 4. Data illustrating the effect of downregulation of the relationships between the yield of charge stabilization at open PS
II centres ($\phi_{S(0)}$), and (*a*) the yield of charge separation (ϕ_C), (*b*) the yield Figure 4. Data illustrating the effect of downregulation of the relationships between the yield of charge stabilization at open PS
II centres ($\phi_{S(0)}$), and (*a*) the yield of charge separation (ϕ_C), (*b*) the yield II centres ($\phi_{S(o)}$), and (*a*) the yield of charge separation (ϕ_C), (*b*) the yield of chlorophyll fluorescence (ϕ_F), (*c*) the yield of charge recombination that leads to formation of ¹P₆₈₀ (ϕ_R), and (*d*) recombination that leads to formation of ¹P₆₈₀ (ϕ_R), and (d) the yield of charge stabilization at closed PS II centres ($\phi_{S(c)}$). The open circles show the relationships at the observed level of downregulation, wh

recombination will only be significant if the yield of charge separation (ϕ_C) is very high, relative to the yield of charge stabilization at open centres (ϕ_{max}) recombination will only be significant if the charge separation (ϕ_C) is very high, relative to of charge stabilization at open centres ($\phi_{S(0)}$).
Anderson *et al.* (1998) have suggested that arge separation (ϕ_C) is very high, relative to the yield
charge stabilization at open centres $(\phi_{S(0)})$.
Anderson *et al.* (1998) have suggested that repeated
peration of P_{++}^+ , at closed centres (through repeated

Anderson *et al.* (1998) have suggested that repeated
generation of P_{680}^+ at closed centres (through repeated
generation of the radical pair) increases the probability
that an 'inadvertent oxidation causes photodamag generation of P_{680}^+ at closed centres (through repeated
generation of the radical pair) increases the probability
that an 'inadvertent oxidation causes photodamage'.
Although the RBP model allows for the yield of $P^$ generation of the radical pair) increases the probability
that an 'inadvertent oxidation causes photodamage'.
Although the RRP model allows for the yield of P_{680}^{+} to
exceed a value of 1 none of the data analysed wit that an 'inadvertent oxidation causes photodamage'.
Although the RRP model allows for the yield of P_{680}^+ to exceed a value of 1, none of the data analysed within this study gave rise to values for the ratio of ϕ_0 Although the RRP model allows for the yield of P_{680}^{+} to exceed a value of 1, none of the data analysed within this study gave rise to values for the ratio of ϕ_C to $\phi_{S(0)}$ of more than 3. Since the lifetime of exceed a value of 1, none of the data analysed within this
study gave rise to values for the ratio of ϕ_C to $\phi_{S(0)}$ of
more than 3. Since the lifetime of P_{680}^+ is some 30 times
longer at centres that have underg study gave rise to values for the ratio of ϕ_C to $\phi_{S(0)}$ of
more than 3. Since the lifetime of P_{680}^+ is some 30 times
longer at centres that have undergone charge stabil-
ization rather than charge recombination more than 3. Since the lifetime of P_{680}^+ is some 30 times
longer at centres that have undergone charge stabil-
ization, rather than charge recombination, even a ratio as
high as 3 for ϕ , to ϕ means that the int longer at centres that have undergone charge stabilization, rather than charge recombination, even a ratio as high as 3 for ϕ_C to $\phi_{S(0)}$ means that the integrated lifetime of P_{\perp}^+ , is almost entirely defined b ization, rather than charge recombination, even a ratio as
high as 3 for ϕ_C to $\phi_{S(0)}$ means that the integrated lifetime
of P_{680}^+ is almost entirely defined by charge stabilization at
onen centres. The data in \rightarrow of P_{680}^{+} is almost entirely defined by charge stabilization at **r** open centres. The data in figure 3 show the range of of P_{680}^{+} is almost entirely defined by charge stabilization at
open centres. The data in figure 3 show the range of
values for $\phi_{S(0)}$ from the fluorescence trace in figure 2.
The large range of these values from open centres. The data in figure 3 show the range of values for $\phi_{S(0)}$ from the fluorescence trace in figure 2.
The large range of these values, from 0.98 in the dark-
adapted state to 0.11 at 2740 umol m⁻²s⁻¹ is n values for $\phi_{S(0)}$ from the fluorescence trace in figure 2.
The large range of these values, from 0.98 in the dark-
adapted state to 0.11 at 2740 µmol m⁻²s⁻¹, is not easy to
reconcile, with the light-dose response o s^{-1} , is The large range of these values, from 0.98 in the dark-
adapted state to 0.11 at $2740 \,\mu\text{mol m}^{-2} \text{s}^{-1}$, is not easy to
reconcile with the light-dose response of photo-
inactivation inactivation. $\frac{1}{100}$ is formed through charge separation at both
Since P_{680}^+ is formed through charge separation at both
then and closed centres, the most important relationship

open and closed centres, the most important relationship within the light-harvesting system of PS II can still result
for assessing the effect of downregulation on the inte-
in the ${}^{1}O_{2}$ -induced photodestruction of c Since P_{680}^+ is formed through charge separation at both
open and closed centres, the most important relationship
for assessing the effect of downregulation on the inte-
grated lifetime of P^+ is that between the vi open and closed centres, the most important relationship
for assessing the effect of downregulation on the inte-
grated lifetime of P_{680}^{+} is that between the yield of charge
stabilization at open centres (ϕ_{ext}) for assessing the effect of downregulation on the integrated lifetime of P_{680}^+ is that between the yield of charge stabilization at open centres ($\phi_{S(0)}$) and the total yield of charge separation (at open plus clos grated lifetime of P_{680}^{+} is that between the yield of charge
stabilization at open centres $(\phi_{S(0)})$ and the total yield of
charge separation (at open plus closed centres), which is
given by ϕ . This relationship stabilization at open centres $(\phi_{S(0)})$ and the total yield of charge separation (at open plus closed centres), which is given by ϕ_C . This relationship is illustrated by the data in figure 4*a*, which are derived from charge separation (at open plus closed centres), which is
given by ϕ_C . This relationship is illustrated by the data in
figure 4*a*, which are derived from the fluorescence trace
in figure 2. The open circles show the r given by ϕ_C . This relationship is illustrated by the data in figure 4*a*, which are derived from the fluorescence trace in figure 2. The open circles show the relationship when [SV] and [O] are adjusted to give the ob figure 4*a*, which are derived from the fluorescence trace
in figure 2. The open circles show the relationship when
[SV] and [Q_A] are adjusted to give the observed values [SV] and $[Q_A]$ are adjusted to give the observed values *Phil. Trans. R. Soc. Lond.* B (2000)

of charge stabilization at open centres $(\phi_{S(0)})$.

Anderson *et al.* (1998) have suggested that repeated by the value of $[Q_A]$ alone. Although the removal of generation of P_{680}^+ at closed centres (through repeated of F_v'/F_m' and F_q'/F_v' while the closed squares show what
the relationship would be if there were no downregulaof F_v'/F_m' and F_q'/F_v' while the closed squares show what
the relationship would be if there were no downregula-
tion ([SV] = 0) and the same values of ϕ_{max} were defined of F_v'/F_m' and F_q'/F_v' while the closed squares show what
the relationship would be if there were no downregula-
tion ([SV] = 0) and the same values of $\phi_{S(s)}$ were defined
by the value of [O, l, alone, Although the re the relationship would be if there were no downregulation ([SV] = 0) and the same values of $\phi_{S(0)}$ were defined
by the value of [Q_A] alone. Although the removal of
downregulation increases the ratio of ϕ_{τ} to $\$ tion ([SV] = 0) and the same values of $\phi_{S(0)}$ were defined
by the value of [Q_A] alone. Although the removal of
downregulation increases the ratio of ϕ_C to $\phi_{S(0)}$ from less
than 2 to over 10 at high incident PPE than 2 to over 10, at high incident PPFD, the much longer lifetime of P_{680}^{+} at open centres where charge stabithan 2 to over 10, at high incident PPFD, the much
longer lifetime of P_{680}^+ at open centres where charge stabi-
lization has occurred still leaves the integrated lifetime of
 P^+ largely defined by ϕ . Consequentl P_{680}^{+} largely defined by $\phi_{S(0)}$. Consequently, downregula-In lifetime of P_{680}^{+} at open centres where charge stabion has occurred still leaves the integrated lifetime of largely defined by $\phi_{S(0)}$. Consequently, downregula-
does not significantly increase ϕ_{max} relat lization has occurred still leaves the integrated lifetime of P_{680}^+ largely defined by $\phi_{S(0)}$. Consequently, downregulation does not significantly increase $\phi_{S(0)}$, relative to the integrated vield of P_{loc}^+ P_{680}^{+} largely defined by $\phi_{S(0)}$. Consequently, downregulation does not significantly increase $\phi_{S(0)}$, relative to the integrated yield of P_{680}^{+} , as would be expected if P_{680}^{+} were a significant tri

(b) ${}^3P_{680}^*$ as a potential trigger of photoinactivation (i) ${}^{3}P_{680}^{*}$ *formed through intersystem crossing formed through intersystem crossing*
formed through intersystem crossing
and in \$1, there are two ways in

As noted in §1, there are two ways in which ${}^{3}P_{680}$ can (i) ${}^{3}P_{680}^{*}$ *formed through intersystem crossing*
As noted in § 1, there are two ways in which ${}^{3}P_{680}$ can
be formed; directly from ${}^{1}P_{680}^{*}$, through intersystem
crossing and through charge recombinati As noted in § 1, there are two ways in which ³P₆₈₀ can
be formed; directly from ¹P₆₈₀, through intersystem
crossing, and through charge recombination between P₆₈₀
and Phe⁻. Both pathways are considered here.
D crossing, and through charge recombination between P_{680}^+
and Phe⁻. Both pathways are considered here.
Despite the protection afforded by the presence of β -

carotene and the putative O_2 barrier, formation of ³Chl^{*}
within the light-harvesting system of PS II can still result
in the ¹O₂-induced photodestruction of chlorophylls, with
an apparent yield of between 10^{-5 within the light-harvesting system of PS II can still result
in the ¹O₂-induced photodestruction of chlorophylls, with
an apparent yield of between 10^{-5} and 10^{-6} (Krasnovsky in the ¹O₂-induced photodestruction of chlorophylls, with
an apparent yield of between 10^{-5} and 10^{-6} (Krasnovsky
1994). If P₆₈₀ is simply considered as one of a number of
chlorophylls within the nigment hed wi an apparent yield of between 10^{-5} and 10^{-6} (Krasnovsky
1994). If P_{680} is simply considered as one of a number of
chlorophylls within the pigment bed, with no more and
no less susceptibility to ¹O induced photo 1994). If P_{680} is simply considere
chlorophylls within the pigment
no less susceptibility to ${}^{1}O_{2}$ -in
than any other then an overall y ered as one of a number of
nt bed, with no more and
-induced photodestruction
ll vield of between 10⁵ and chlorophylls within the pigment bed, with no more and
no less susceptibility to ${}^{1}O_{2}$ -induced photodestruction
than any other, then an overall yield of between 10^{5} and
 10^{6} would be at least an order of magnit no less susceptibility to ¹O₂-induced photodestruction
than any other, then an overall yield of between 10^5 and
 10^6 would be at least an order of magnitude too low to be
considered as a potential trigger for pho than any other, then an overall yield of between 10^5 and 10^6 would be at least an order of magnitude too low to be considered as a potential trigger for photoinactivation.

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higher than the average
pigment bed as a whole.
First, the distribution First, then the average for chlorophylls within the
gment bed as a whole.
First, the distribution of excitation energy within the
general bed although not strictly a Bolzmann distribu-

pigment bed as a whole.
First, the distribution of excitation energy within the
pigment bed, although not strictly a Bolzmann distribu-
tion is still largely dependent on the spectral characteris-First, the distribution of excitation energy within the pigment bed, although not strictly a Bolzmann distribution, is still largely dependent on the spectral characteristics of the chlorophylls present (Laible *et al.* 1 pigment bed, although not strictly a Bolzmann distribution, is still largely dependent on the spectral characteristics of the chlorophylls present (Laible *et al.* 1994; Dau & the Sauer 1996) Consequently the rapid equilib tion, is still largely dependent on the spectral characteristics of the chlorophylls present (Laible *et al.* 1994; Dau & Sauer 1996). Consequently, the rapid equilibration of excitics of the chlorophylls present (Laible *et al.* 1994; Dau & Sauer 1996). Consequently, the rapid equilibration of excitation energy within the pigment bed results in a higher density of excitation energy on the longer w Sauer 1996). Consequently, the rapid equilibration of excitation energy within the pigment bed results in a higher density of excitation energy on the longer wavelength chlorophylls, which includes P. Second, the potential tation energy within the pigment bed results in a higher
density of excitation energy on the longer wavelength
chlorophylls, which includes P_{680} . Second, the potential
lack of a proximal B-carotene could increase the density of excitation energy on the longer wavelength
chlorophylls, which includes P_{680} . Second, the potential
lack of a proximal β -carotene could increase the lifetime chlorophylls, which includes P_{680} . Second, the potential lack of a proximal β -carotene could increase the lifetime of ${}^{3}P_{680}^{*}$ by orders of magnitude, thereby increasing the vield and lifetime of ${}^{1}\Omega$. lack of a proximal β -carot
of ${}^{3}P_{680}^{*}$ by orders of mag
yield and lifetime of ${}^{1}O_{2}$.
There is no obvious reas ${}^{3}P_{680}^{*}$ by orders of magnitude, thereby increasing the lead and lifetime of ${}^{1}Q_{2}$.
There is no obvious reason why a centre being open or order than the probability

yield and lifetime of 1O_2 .
There is no obvious reason why a centre being open or closed should have any impact on either the probability of an exciton 'visiting' P are the vield of ${}^3P^*$ that is There is no obvious reason why a centre being open or closed should have any impact on either the probability of an exciton 'visiting' P_{680} or the yield of ${}^{3}P_{680}^{*}$ that is formed through intersystem crossing. closed should have any impact on either the probability
of an exciton 'visiting' P_{680} or the yield of ${}^{3}P_{680}^{*}$ that is
formed through intersystem crossing. Consequently, the
vield of ${}^{3}P_{\infty}^{*}$, formation of an exciton 'visiting' P_{680} or the yield of ${}^{3}P_{680}^{*}$ that is
formed through intersystem crossing. Consequently, the
yield of ${}^{3}P_{680}^{*}$ formation through intersystem crossing is
largely dependent on the formed through intersystem crossing. Consequently, the yield of ${}^{3}P_{680}^{*}$ formation through intersystem crossing is largely dependent on the lifetime of an exciton, which is proportional to the vield of fluorescence yield of ³P₆₈₀ formation through intersystem creasurely dependent on the lifetime of an exciton, proportional to the yield of fluorescence (ϕ_F) . Solven the yield of fluorescence (ϕ_F) . yield of ³P₆₈₀ formation through intersystem crossing is
largely dependent on the lifetime of an exciton, which is
proportional to the yield of fluorescence (ϕ_F). Since ϕ_F
changes very little with PPFD (see figu proportional to the yield of fluorescence (ϕ_F) . Since ϕ_F changes very little with PPFD (see figure 4*b*), the concept of photoinactivation being triggered by formation of ³P₆₈₀ through intersystem crossing fits w changes very little with PPFD (see figure 4b), the concept
of photoinactivation being triggered by formation of ${}^{3}P_{680}^{*}$
through intersystem crossing fits well with the light-dose
response of photoinactivation of photoinactivation being trigge
through intersystem crossing fit
response of photoinactivation.
One issue that needs to be a through intersystem crossing fits well with the light-dose
response of photoinactivation.
One issue that needs to be addressed is the process of

response of photoinactivation.
One issue that needs to be addressed is the process of
charge separation and recombination that results in refor-
mation of \mathbb{P}^* since this accounts for a fraction of the One issue that needs to be addressed is the process of charge separation and recombination that results in reformation of ${}^{1}P_{680}^{*}$, since this accounts for a fraction of the lifetime of an exciton. The relationship charge separation and recombination that results in reformation of ${}^{1}P_{680}^{*}$, since this accounts for a fraction of the lifetime of an exciton. The relationship between $\phi_{S(0)}$ and the vield of charge recombinatio mation of ¹P₆₈₀, since this accounts for a fraction of the lifetime of an exciton. The relationship between $\phi_{S(0)}$ and the yield of charge recombination (ϕ_R), derived from the lifetime of an exciton. The relationship between $\phi_{S(o)}$ and
the yield of charge recombination (ϕ_R) , derived from the
trace in figure 2, is shown in figure 4*c*. These data show
that ϕ_r varies by less than a factor the yield of charge recombination (ϕ_R) , derived from the trace in figure 2, is shown in figure 4c. These data show that ϕ_R varies by less than a factor of 2 and has a maximum value (at the lowest PPEDs) of slightly o trace in figure 2, is shown in figure 4c. These data show
that ϕ_R varies by less than a factor of 2 and has a
maximum value (at the lowest PPFDs) of slightly over
0.1. The time taken for reformation of \mathbb{P}_{++}^* thr that ϕ_R varies by less than a factor of 2 and has a maximum value (at the lowest PPFDs) of slightly over 0.1. The time taken for reformation of ${}^{1}P_{680}^{*}$ through charge separation and recombination is in the same maximum value (at the lowest PPFDs) of slightly over
0.1. The time taken for reformation of ${}^{1}P_{680}^{*}$ through
charge separation and recombination is in the same range
as the mean lifetime of an exciton within the ni 0.1. The time taken for reformation of ${}^{1}P_{680}^{*}$ through charge separation and recombination is in the same range as the mean lifetime of an exciton within the pigment bed (Roelofs *et al.* 1992) and consequently do charge separation and recombination is in the same range
as the mean lifetime of an exciton within the pigment bed
(Roelofs *et al.* 1992) and, consequently, does not represent as the mean lifetime of an exciton within the pigment bed (Roelofs *et al.* 1992) and, consequently, does not represent a strong argument against ${}^3P_{680}^*$ being a trigger for photo-inactivation inactivation. trong argument against ${}^3P_{680}^*$ being a trigger for photo-
activation.
The effect of downregulation on the yield of ${}^3P_{680}^*$
mation through intersustem crossing can be assessed

The effect of downregulation on the yield of ${}^{3}P_{680}^{*}$
formation through intersystem crossing can be assessed
from the data in figure $4b$,*c*. As with the data in figure $4a$,
the impact of downregulation was asses formation through intersystem crossing can be assessed
from the data in figure $4b$, c . As with the data in figure $4a$,
the impact of downregulation was assessed by setting
 $[SN]$ to zero in the modelled data, and achie from the data in figure $4b$, c . As with the data in figure $4a$, the impact of downregulation was assessed by setting [SV] to zero in the modelled data, and achieving the observed value of F'/F' by adjusting [O]. It the impact of downregulation was assessed by setting [SV] to zero in the modelled data, and achieving the observed value of F'_q/F'_m by adjusting [Q_A]. It is clear, from these data, that downregulation decreases both $\$ [SV] to zero in the modelled data, and achieving the observed value of F'_q/F'_m by adjusting [Q_A]. It is clear, from these data, that downregulation decreases both ϕ_F and ϕ_R in roughly equal proportion and would, from these data, that downregulation decreases both ϕ_F and ϕ_R in roughly equal proportion and would, therefore, be expected to decrease the yield of ${}^{3}P_{680}^{*}$ formation and ϕ_R in roughly equal propo
be expected to decrease the
through intersystem crossing. through intersystem crossing.
(ii)³*P*₆₈₀ *formed through charge recombination*
 $\Lambda_{6.0}$ acted above (8.2(a)), the formation

 $A^{3}P_{680}$ *formed through charge recombination*
As noted above (§ 3(a)), the formation of ${}^{3}P_{680}^{*}$ there recombination is one of three processes As noted above (§ 3(a)), the formation of ${}^{3}P_{680}^{*}$ through (ii)³ P_{680} formed through charge recombination
As noted above (§ 3(a)), the formation of ³ P_{680}^* through
charge recombination is one of three processes that
contribute to charge stabilization at open and closed As noted above (§ 3(a)), the formation of ${}^{3}P_{680}^{*}$ through
charge recombination is one of three processes that
contribute to charge stabilization at open and closed
centres the other two being formation of ¹P thr charge recombination is one of three processes that
contribute to charge stabilization at open and closed
centres, the other two being formation of ${}^{1}P_{680}$ through
charge recombination and charge stabilization result contribute to charge stabilization at open and closed formation of ${}^{3}P_{680}^{*}$ through charge recombination and
centres, the other two being formation of ${}^{1}P_{680}$ through double reduction of Q_A) could be reconci centres, the other two being formation of ¹P₆₈₀ through
charge recombination and charge stabilization, resulting
from electron transfer to Q_A . Consequently, the yield of
³P₆₈₀ through charge recombination cannot directly. ${}^{3}P_{680}^{*}$ through charge recombination cannot be calculated directly.
If it is assumed that the probability of charge recombi-

nation leading to the formation of ${}^{3}P_{680}^{*}$ (rather than ${}^{1}P_{680}$ or ${}^{1}P_{680}^{*}$) is the same at open and closed centres, then the

g of *photoinactivation* K. Oxborough and N. R. Baker 1495
yield of ${}^{3}P_{680}^{*}$ would obviously be proportional to the yield yield of ${}^{3}P_{680}^{*}$ would obviously be proportional to the yield
of ${}^{1}P_{680}^{*}$ given by ϕ_{R} , which can be calculated. The rela-
tionship between ϕ_{max} and ϕ_{R} for the trace in figure 2 is yield of ³P₆₈₀ would obviously be proportional to the yield
of ¹P₆₈₀, given by ϕ_R , which can be calculated. The rela-
tionship between $\phi_{S(0)}$ and ϕ_R for the trace in figure 2 is
demonstrated by the data i of ¹P₆₈₀, given by ϕ_R , which can be calculated. The rela-
tionship between $\phi_{S(0)}$ and ϕ_R for the trace in figure 2 is
demonstrated by the data in figure 4*c*, which show a 44%
decrease in ϕ - between the hi tionship between $\phi_{S(0)}$ and ϕ_R for the trace in figure 2 is
demonstrated by the data in figure 4c, which show a 44%
decrease in ϕ_R between the highest and lowest values.
Although strict compatibility with the lig demonstrated by the data in figure 4c, which show a 44% decrease in ϕ_R between the hightest and lowest values.
Although strict compatibility with the light-dose response would require a stable yield for ϕ_R with chan decrease in ϕ_R between the highest and lowest values. the relatively narrow range observed is not incompatible would require a stable yield for ϕ_R with changing $\phi_{S(o)}$
the relatively narrow range observed is not incompatible
with ${}^3P_{680}^*$ formed through charge recombination being a
trigger for photoinactivation the relatively narrow range ob
with ${}^{3}P_{680}^{*}$ formed through cha
trigger for photoinactivation.
As already noted, the data th ${}^{3}P_{680}^{*}$ formed through charge recombination being a
gger for photoinactivation.
As already noted, the data in figure 4*c* show that
write universal decreases ϕ , with the largest effect being

trigger for photoinactivation.
As already noted, the data in figure 4ϵ show that
downregulation decreases ϕ_R , with the largest effect being
seen at the highest PPFD values (lowest values of ϕ_{max}) As already noted, the data in figure $4c$ show that
downregulation decreases ϕ_R , with the largest effect being
seen at the highest PPFD values (lowest values of $\phi_{S(0)}$).
Consequently, downregulation would be expecte downregulation decreases ϕ_R , with the largest effect being
seen at the highest PPFD values (lowest values of $\phi_{S(0)}$).
Consequently, downregulation would be expected to seen at the highest PPFD values (lowest values of $\phi_{S(0)}$).
Consequently, downregulation would be expected to decrease the yields of ${}^{3}P_{680}^{*}$ formed through either inter-
system crossing or charge recombination Consequently, downregulation would be
decrease the yields of ${}^{3}P_{680}^{*}$ formed throug
system crossing or charge recombination.

(c) *Double reduction of* Q_A *as a potential trigger*
(c) *Double reduction of* Q_A *as a potential trigger of photoinactivation*

inactivation.

The effect of downregulation on the yield of ${}^{3}P_{680}^{*}$ that is consistent with the lower yield at low PPFDs

formation through intersystem crossing can be assessed observed here. The data in figure 4 **of photoinactivation**
The possibility that double reduction of Q_A could be a **of photoinactivation**
The possibility that double reduction of Q_A could be a
trigger for photoinactivation (Van Wijk *et al.* 1992; Vass *et*
 a^{l} 1992) has been strongly contested on the grounds that The possibility that double reduction of Q_A could be a trigger for photoinactivation (Van Wijk *et al.* 1992; Vass *et al.* 1992) has been strongly contested on the grounds that the application of target theory reveals trigger for photoinactivation (Van Wijk *et al.* 1992; Vass *et al.* 1992) has been strongly contested on the grounds that the application of target theory reveals that photoinactivation is a single-photon event (Sinclair al. 1992) has been strongly contested on the grounds that
the application of target theory reveals that photoinacti-
vation is a single-photon event (Sinclair *et al.* 1996;
Anderson *et al.* 1998) Whilet double reduction the application of target theory reveals that photoinactivation is a single-photon event (Sinclair *et al.* 1996; Anderson *et al.* 1998). Whilst double reduction of Q_A at onen PS II centres is clearly a two-photon even vation is a single-photon event (Sinclair *et al.* 1996;
Anderson *et al.* 1998). Whilst double reduction of Q_A at
open PS II centres is clearly a two-photon event, it is
perfectly valid to consider closed PS II centres Anderson *et al.* 1998). Whilst double reduction of Q_A at open PS II centres is clearly a two-photon event, it is perfectly valid to consider closed PS II centres as targets, open PS II centres is clearly a two-photon event, it is
perfectly valid to consider closed PS II centres as targets,
where double reduction of Q_A is actually a single-photon
event (since, by definition Q_A already carr perfectly valid to consider closed PS II centres as targets,
where double reduction of Q_A is actually a single-photon
event (since, by definition, Q_A already carries a single
perspitue charge at these centres). The ob where double reduction of Q_A is actually a single-photon
event (since, by definition, Q_A already carries a single
negative charge at these centres). The observation that
photoinactivation is a single-photon event can event (since, by definition, Q_A already carries a single negative charge at these centres). The observation that photoinactivation is a single-photon event can be reconnegative charge at these centres). The observation that
photoinactivation is a single-photon event can be recon-
ciled with closed centres being the target, providing the
probability of a photon-inducing stable charge sepa photoinactivation is a single-photon event can be reconciled with closed centres being the target, providing the probability of a photon-inducing stable charge separation at a closed centre is insensitive to PPED. The data ciled with closed centres being the target, providing the
probability of a photon-inducing stable charge separation
at a closed centre is insensitive to PPFD. The data in
figure 4d show that the probability of a photon bei probability of a photon-inducing stable charge separation at a closed centre is insensitive to PPFD. The data in figure $4d$ show that the probability of a photon being used to drive stable charge separation at a closed centre increases with light at low PPFD, but remains fairly figure $4d$ show that the probability of a photon being used
to drive stable charge separation at a closed centre
increases with light at low PPFD, but remains fairly
stable as PPFD increases beyond this range. Sinclair to drive stable charge separation at a closed centre increases with light at low PPFD, but remains fairly
stable as PPFD increases beyond this range. Sinclair *et al.*
(1996) noted that the yield of photoinactivation is low at
low photon doses, which included low PPFDs: a re stable as PPFD increases beyond this range. Sinclair *et al.* (1996) noted that the yield of photoinactivation is low at low photon doses, which included low PPFDs; a result that is consistent with the lower yield at low P (1996) noted that the yield of photoinactivation is low at
low photon doses, which included low PPFDs; a result
that is consistent with the lower yield at low PPFDs
observed here. The data in figure $4d$ also show that wi observed here. The data in ¢gure 4*^d* also show that, with that is consistent with the lower yield at low PPFDs
observed here. The data in figure 4d also show that, with
increasing PPFD (going from right to left along the
reavis) downregulation increases the ratio of ϕ_{max} t observed here. The data in figure $4d$ also show that, with increasing PPFD (going from right to left along the *x*-axis), downregulation increases the ratio of $\phi_{S(0)}$ to $\phi_{S(c)}$ and therefore provides a bigh level of increasing PPFD (going from right to left along the *x*-axis), downregulation increases the ratio of $\phi_{S(0)}$ to $\phi_{S(c)}$ and therefore provides a high level of protection against double reduction of Ω : a result that *x*-axis), downregulation increases the ratio of $\phi_{S(0)}$ to $\phi_{S(c)}$ and therefore provides a high level of protection against double reduction of Q_A ; a result that is also consistent with triggering of photoinactivat and therefore provides a high level of protection against

4. DISCUSSION

The objective of this study was to determine how well four potential triggers of photoinactivation (formation of The objective of this study was to determine how well
four potential triggers of photoinactivation (formation of
 P_{680}^+ , formation of ${}^{3}P_{680}^*$ through intersystem crossing,
formation of ${}^{3}P_{60}^*$, through ch four potential triggers of photoinactivation (formation of P_{680}^+ , formation of ${}^3P_{680}^*$ through intersystem crossing,
formation of ${}^3P_{680}^*$ through charge recombination and
double reduction of Ω) could P_{680}^{+} , formation of ${}^{3}P_{680}^{*}$ through intersystem crossing,
formation of ${}^{3}P_{680}^{*}$ through charge recombination and
double reduction of Q_A) could be reconciled with the
reversible exciton-radical pair formation of ${}^{3}P_{680}^{*}$ through charge recombination and
double reduction of Q_A) could be reconciled with the
reversible exciton-radical pair equilibrium model of
Schatz et al. (1988) a Stern-Volmer model for downdouble reduction of Q_A) could be reconciled with the reversible exciton-radical pair equilibrium model of Schatz *et al.* (1988), a Stern-Volmer model for down-regulation and the apparent light-dose response of photoreversible exciton-radical pair equilibrium model of Schatz *et al.* (1988), a Stern-Volmer model for down-regulation and the apparent light-dose response of photo-inactivation inactivation. It has been argued that the apparent light-dose response of photo-
It has been argued that the apparent light-dose
propes of photoinactivation is strong evidence for a

inactivation.
It has been argued that the apparent light-dose
response of photoinactivation is strong evidence for a
single trigger (Anderson *et al*, 1998) Although it would It has been argued that the apparent light-dose
response of photoinactivation is strong evidence for a
single trigger (Anderson *et al.* 1998). Although it would

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perhaps be surprising if two or more triggering processes had similar yields (and were, therefore, both significant perhaps be surprising if two or more triggering processes
had similar yields (and were, therefore, both significant
triggers of the photoinactivation process), the apparent
light-dose response of photoinactivation is not i had similar yields (and were, therefore, both significant
triggers of the photoinactivation process), the apparent
light-dose response of photoinactivation is not, in itself,
an argument against this being the case. One cr triggers of the photoinactivation process), the apparent
light-dose response of photoinactivation is not, in itself,
an argument against this being the case. One criterion
that does need to be satisfied is that the vield o light-dose response of photoinactivation is not, in itself, an argument against this being the case. One criterion that does need to be satisfied is that the yield of any an argument against this being the case. One criterion that does need to be satisfied is that the yield of any putative trigger should be constant with changing PPFD. Given the strong evidence that downregulation 'protects putative trigger should be constant with changing PPFD. against photoinactivation, it seems reasonable to specify Given the strong evidence that downregulation 'protects'
against photoinactivation, it seems reasonable to specify
a second criterion for any potential trigger of photo-
inactivation: that its vield is lowered by this proc against photoinactivation, it seems reasonable to specify
a second criterion for any potential trigger of photo-
inactivation; that its yield is lowered by this process,
relative to ϕ . When these two criteria are consi a second criterion for any potential trigger of photo-
inactivation; that its yield is lowered by this process,
relative to $\phi_{C(o)}$. When these two criteria are considered
together, the formation of ${}^{3}P_{++}^{*}$ through inactivation; that its yield is lowered by this process,
relative to $\phi_{C(o)}$. When these two criteria are considered through intersystem by
together, the formation of ${}^{3}P_{680}^{*}$ through intersystem by
crossing is pe relative to $\phi_{C(o)}$. When these two criteria are considered

dependence together, the formation of ${}^3P_{680}^*$ through intersystem

dependence of the strongest candidate, followed by together, the formation of ${}^{3}P_{680}^{*}$ through intersystem
crossing is perhaps the strongest candidate, followed by
formation of ${}^{3}P_{680}^{*}$ through charge recombination and
double reduction of O_{\perp} . Formation crossing is perhaps the strongest candidate, followed by
formation of ${}^{3}P_{680}^{*}$ through charge recombination and
double reduction of Q_A . Formation of P_{680}^{*} is a poor
candidate in the context of both criteria formation of ${}^{3}P_{680}^{*}$ through charge recombination and
double reduction of Q_A . Formation of P_{680}^{*} is a poor
candidate in the context of both criteria, once the large
difference between the lifetimes of P^{+ double reduction of Q_A . Formation of P_{680}^+ is a poor
candidate in the context of both criteria, once the large
difference between the lifetimes of P_{680}^+ that is taken
hack to the ground state through electron candidate in the context of both criteria, once the large
difference between the lifetimes of P_{680}^{+} that is taken
back to the ground state through electron transfer from
Z and through charge recombination with Phe⁻ difference between the lifetimes of P_{680}^{+} that is taken
back to the ground state through electron transfer from
Z and through charge recombination with Phe[–] is taken
into account back to the gree
Z and through
into account.

and through charge recombination with Phe⁻ is taken
to account.
A third criterion that obviously needs to be satisfied is
at the vield of any putative trigger should be high into account.
A third criterion that obviously needs to be satisfied is
that the yield of any putative trigger should be high
enough to account for the observed yield of photo-A third criterion that obviously needs to be satisfied is
that the yield of any putative trigger should be high
enough to account for the observed yield of photo-
inactivation, estimated at between 10^{-6} and 10^{-7} by that the yield of any putative trigger should be high
enough to account for the observed yield of photo-
inactivation, estimated at between 10^{-6} and 10^{-7} by
Anderson *et al.* (1997) From the information currently enough to account for the observed yield of photo-
inactivation, estimated at between 10^{-6} and 10^{-7} by
Anderson *et al.* (1997). From the information currently
available it would seem that the vield of ${}^{3}P_{\infty}^{*$ inactivation, estimated at between 10^{-6} and 10^{-7} by
Anderson *et al.* (1997). From the information currently
available, it would seem that the yield of ${}^{3}P_{680}^{*}$ through
intersystem crossing is likely to be be Anderson *et al.* (1997). From the information currently available, it would seem that the yield of ${}^{3}P_{680}^{*}$ through intersystem crossing is likely to be between 10^{-3} and 10^{-4} assuming that its vield is rough 10^{-4} , assuming that its yield is roughly twice that of chlorophyll fluorescence (Durrant *et al.* 1990). Whether or not this is high enough for it to be a serious contender largely denends of the level of protection th lable, it would seem that the yield of ${}^{3}P_{680}^{*}$ through
system crossing is likely to be between 10^{-3} and
is assuming that its yield is roughly twice that of
coppell fluorescence (Durrant et al. 1990) Whether or intersystem crossing is likely to be between 10^{-3} and 10^{-4} , assuming that its yield is roughly twice that of chlorophyll fluorescence (Durrant *et al.* 1990). Whether or not this is high enough for it to be a seriou chlorophyll fluorescence (Durrant *et al.* 1990). Whether or
not this is high enough for it to be a serious contender
largely depends of the level of protection that is afforded by
 β -carotene. If this protection is as e not this is high enough for it to be a serious contender
largely depends of the level of protection that is afforded by
 β -carotene. If this protection is as efficient as that afforded
the other chlorophylls within the largely depends of the level of protection that is afforded by
 β -carotene. If this protection is as efficient as that afforded
the other chlorophylls within the pigment bed of PS II, then
the vield of ${}^{3}P_{\infty}^{*}$, β -carotene. If this protection is as efficient as that afforded
the other chlorophylls within the pigment bed of PS II, then
the yield of ${}^{3}P_{680}^{*}$ through intersystem crossing would be
orders of magnitude too lo the yield of ${}^{3}P_{680}^{*}$ through intersystem crossing would be the other chlorophylls within the pigment bed of PS II, then
the yield of ${}^{3}P_{680}^{*}$ through intersystem crossing would be
orders of magnitude too low, since its lifetime would prob-
ably be too short for it to induc the yield of ${}^{3}P_{680}^{*}$ through intersystem crossing would be
orders of magnitude too low, since its lifetime would prob-
ably be too short for it to induce significant formation of
 ${}^{1}O_{2}$. However, the lifetime ably be too short for it to induce significant formation of 1O_2 . However, the lifetime of ${}^3P_{680}$ could be increased by orders of magnitude if, as seems perfectly feasible, the level of protection afforded by $R_{$ ¹O₂. However, the lifetime of ³P₆₈₀ could be increased by orders of magnitude if, as seems perfectly feasible, the level of protection afforded by β -carotene is very low (Barber 1998) A vield for ³P^{*} of $Ca^{$ orders of magnitude if, as seems perfectly feasible, the level
of protection afforded by β -carotene is very low (Barber
1998). A yield for ${}^{3}P_{680}^{*}$ of *ca.* 30% has been observed at closed centres within isolated reaction centre complexes 1998). A yield for ${}^{3}P_{680}^{*}$ of *ca*. 30% has been observed at closed centres within isolated reaction centre complexes (Durrant *et al.* 1990). Most of this yield was attributed to charge recombination largely beca closed centres within isolated reaction centre complexes
(Durrant *et al.* 1990). Most of this yield was attributed to
charge recombination, largely because the yield of fluores-
cence was only 2% and the yield of ${}^{3}P^$ (Durrant *et al.* 1990). Most of this yield was attributed to charge recombination, largely because the yield of fluorescence was only 2% and the yield of ${}^{3}P_{680}^{*}$ through inter-
system crossing is likely to be rou charge recombination, largely because the yield of fluorescence was only 2% and the yield of ${}^{3}P_{680}^{*}$ through inter-
system crossing is likely to be roughly twice this figure.
These data strongly suggest that the f cence was only 2% and the yield of ${}^{3}P_{680}^{*}$ through inter-
system crossing is likely to be roughly twice this figure.
These data strongly suggest that the formation of ${}^{3}P_{680}^{*}$
through charge recombination i System crossing is likely to be roughly twice this figure.
These data strongly suggest that the formation of ${}^{3}P_{680}^{*}$
through charge recombination is likely to be a much more
efficient trigger of photoinactivation These data strongly suggest that the formation of ${}^{3}P_{680}^{*}$ $\bigcup{}^{3}P_{680}^{*}$ through intersystem crossing.

It has been argued previously that double reduction of ${}^{3}P_{680}^{*}$ through intersystem crossing.
It has been argued previously that double reduction of
 Q_A is unlikely as a mechanism of photoinactivation
because the probability of charge separation occurring at It has been argued previously that double reduction of Q_A is unlikely as a mechanism of photoinactivation because the probability of charge separation occurring at a closed centre is too low (Park *et al.* 1997: Anderso Q_A is unlikely as a mechanism of photoinactivation
because the probability of charge separation occurring at
a closed centre is too low (Park *et al.* 1997; Anderson *et al.*
1998) In actual fact, as the data in figure because the probability of charge separation occurring at
a closed centre is too low (Park *et al.* 1997; Anderson *et al.*
1998). In actual fact, as the data in figure 4*d* clearly
illustrate, the vield of charge stabiliz a closed centre is too low (Park *et al.* 1997; Anderson *et al.* 1998). In actual fact, as the data in figure $4d$ clearly illustrate, the yield of charge stabilization at closed centres is relatively high once PPED is a 1998). In actual fact, as the data in figure $4d$ clearly illustrate, the yield of charge stabilization at closed \perp centres is relatively high, once PPFD is above illustrate, the yield of charge stabilization at closed
centres is relatively high, once PPFD is above
ca. 200 µmol m⁻²s⁻¹, at between 0.06 and 0.16. Although
the fraction of charge stabilization events that result centres is relatively high, once PPFD is above
ca. 200 μ mol m⁻²s⁻¹, at between 0.06 and 0.16. Although
the fraction of charge stabilization events that result in
double reduction of Q is not known a comparison of ca. 200 µmol m⁻²s⁻¹, at between 0.06 and 0.16. Although
the fraction of charge stabilization events that result in
double reduction of Q_A is not known, a comparison of
the vields of photoinactivation (between 10^{-6} the fraction of charge stabilization events that result in
double reduction of Q_A is not known, a comparison of
the yields of photoinactivation (between 10^{-6} and 10^{-7})
and charge stabilization at closed centres (c double reduction of Q_A is not known, a comparison of
the yields of photoinactivation (between 10^{-6} and 10^{-7})
and charge stabilization at closed centres (*ca*. 10^{-1}) clearly
shows that it would not have to be ve the yields of photoinactivation (between 10^{-6}
and charge stabilization at closed centres (*ca*. 1
shows that it would not have to be very high. *Phil. Trans. R. Soc. Lond.* B (2000) *Phil. Trans. R. Soc. Lond.* B (2000)

In conclusion, the formation of ${}^{3}P_{680}^{*}$ through inter-
tem crossing, and through charge recombination and In conclusion, the formation of ${}^{3}P_{680}^{*}$ through inter-
system crossing, and through charge recombination and
the double reduction of O are all feasible as triggers for In conclusion, the formation of ${}^{3}P_{680}^{*}$ through inter-
system crossing, and through charge recombination and
the double reduction of Q_A , are all feasible as triggers for
the photoinactivation process in terms of system crossing, and through charge recombination and
the double reduction of Q_A , are all feasible as triggers for
the photoinactivation process in terms of the first two
above criteria. That is, their yields are relati the double reduction of Q_A , are all feasible as triggers for
the photoinactivation process in terms of the first two
above criteria. That is, their yields are relatively insensi-
tive to PPFD and their yields are lowere the photoinactivation process in terms of the first two above criteria. That is, their yields are relatively insensi-
tive to PPFD and their yields are lowered, relative to
 $\phi_{S(0)}$ by downregulation. P_{680}^+ clearly fails both of these
criteria. Given that the vield of ${}$ tive to PPFD and their yields are lowered, relative to $\phi_{S(0)}$ by downregulation. P_{680}^{+} clearly fails both of these criteria. Given that the yield of ${}^{3}P_{680}^{*}$ formed through charge recombination is likely t $\phi_{S(o)}$ by downregulation. P_{680}^{+} clearly fails both of these criteria. Given that the yield of ${}^{3}P_{680}^{*}$ formed through charge recombination is likely to be much higher than the yield of ${}^{3}P_{\infty}^{*}$, form criteria. Given that the yield of ${}^{3}P_{680}^{*}$ formed through
charge recombination is likely to be much higher than the
yield of ${}^{3}P_{680}^{*}$ formed through intersystem crossing, it
seems unlikely on current evidenc charge recombination is likely to be much higher the yield of ${}^{3}P_{680}^{*}$ formed through intersystem crossi
seems unlikely, on current evidence, that ${}^{3}P_{680}^{*}$ for
through the latter pathway makes a significant seems unlikely, on current evidence, that ${}^{3}P_{680}^{*}$ formed
through the latter pathway makes a significant contri-
bution to the photoinactivation process. The relatively
high yield of charge stabilization at closed yield of ${}^{3}P_{680}^{*}$ formed through intersystem crossing, it
seems unlikely, on current evidence, that ${}^{3}P_{680}^{*}$ formed
through the latter pathway makes a significant contri-
bution to the photoinactivation proc through the latter pathway makes a significant contribution to the photoinactivation process. The relatively
high yield of charge stabilization at closed centres, at all
but the lowest PPEDs, means that double reduction of bution to the photoinactivation process. The relatively
high yield of charge stabilization at closed centres, at all
but the lowest PPFDs, means that double reduction of
O, should also be considered as a viable trigger of high yield of charge stabilization at closed centres, at all
but the lowest PPFDs, means that double reduction of
 Q_A should also be considered as a viable trigger of photo-
inactivation inactivation.

APPENDIX A. EXPLANATION OF ABBREVIATIONS

F', chlorophyll fluorescence signal in the light-adapted F' , chlorophyll fluorescence signal in the light-adapted
state; F_m and F'_m , chlorophyll fluorescence signal when all
PS II centres are closed in the dark, and light-adapted F' , chlorophyll fluorescence signal in the light-adapted
state; F_m and F'_m , chlorophyll fluorescence signal when all
PS II centres are closed in the dark- and light-adapted
states respectively: F and F' chloroph state; F_m and F'_m , chlorophyll fluorescence signal when all
PS II centres are closed in the dark- and light-adapted
states, respectively; F_o and F'_o , chlorophyll fluorescence
signal when all centres are open in th PS II centres are closed in the dark- and light-adapted states, respectively; F_0 and F'_0 , chlorophyll fluorescence signal when all centres are open in the dark- and lightstates, respectively; F_0 and F'_0 , chlorophyll fluorescence
signal when all centres are open in the dark- and light-
adapted states, respectively; F'_0 difference between F' and
 $F' \cdot F / F$ and F' / F' fluorescence, *F* is signal when all centres are open in the dark- and light-
adapted states, respectively; $F'_{\rm q}$, difference between F' and
 $F'_{\rm m}$; $F_{\rm v}/F_{\rm m}$ and $F'_{\rm v}/F'_{\rm m}$, fluorescence parameter that
provides an adapted states, respectively; $F'_{\rm q}$, difference between F' and $F'_{\rm w}$; $F_{\rm v}/F_{\rm m}$ and $F'_{\rm v}/F'_{\rm m}$, fluorescence parameter that provides an estimate of the maximum efficiency of PS II photochemistry (when $F'_{\rm m}$; $F_{\rm v}/F_{\rm m}$ and $F'_{\rm v}/F'_{\rm m}$, fluorescence parameter that
provides an estimate of the maximum efficiency of PS II
photochemistry (when $[Q_{\rm A}] = 1$) in the dark- and light-
adapted states respectively: provides an estimate of the maximum efficiency of PS II
photochemistry (when $[Q_A] = 1$) in the dark- and light-
adapted states, respectively; F'_q/F'_m , fluorescence para-
meter that provides an estimate of the efficiency photochemistry (when $[Q_A] = 1$) in the dark- and light-
adapted states, respectively; F_q'/F_m' , fluorescence para-
meter that provides an estimate of the efficiency of PS II
photochemistry (the product of F'/F' and F'/F' adapted states, respectively; $F'_{\rm q}/F'_{\rm m}$
meter that provides an estimate of the product of $F'_{\rm v}/F'_{\rm r}$
photochemistry (the product of $F'_{\rm v}/F'_{\rm r}$ $F'_{\rm w}/F'_{\rm m}$ and $F'_{\rm q}/F'_{\rm v}$; $F'_{\rm q}/F'_{\rm v}$, meter that provides an estimate of the efficiency of PS II

photochemistry (the product of F_v'/F_m' and F_q'/F_v'); F_q'/F_v' ,

fluorescence parameter that quantifies the photochemical

capacity of PS II: F' and F' , var photochemistry (the product of F_v'/F_m' and F_q'/F_v'); F_q'/F_v' ,
fluorescence parameter that quantifies the photochemical
capacity of PS II; F_v' and F_v' , variable chlorophyll fluores-
cence $(F - F)$ or $F' - F_v'$; k_v ap capacity of PS II; F'_{v} and F'_{v} , variable chlorophyll fluoresfluorescence parameter that quantifies the photochemical
capacity of PS II; F'_{ν} and F'_{ν} , variable chlorophyll fluores-
cence $(F_m - F_o)$ or $F'_m - F'_o$; k_C , apparent rate constant for
charge separation at PS II: k capacity of PS II; F_v' and F_v' , variable chlorophyll fluorescence $(F_m - F_o)$ or $F'_m - F_o'$; k_C , apparent rate constant for charge separation at PS II; k_D , apparent rate constant for non-radiative decay at PS II in th cence $(F_m - F_o$ or $F'_m - F_o')$; k_C , apparent rate constant for charge separation at PS II; k_D , apparent rate constant for non-radiative decay at PS II in the dark-adapted state; k apparent rate constant for chlorophyll k_{F} , apparent rate constant for chlorophyll *a* fluorescence;
 k_{R} , apparent rate constant for charge recombination at

PS II, leading to formation of ¹P₆₈₀; k_{S} , apparent rate

constant for the sum charge separation at PS II; k_D , apparent rate constant for non-radiative decay at PS II in the dark-adapted state; PS II, leading to formation of ${}^{1}P_{680}^{*}$; k_S , apparent rate constant for the sum of stable charge separation and $k_{\rm R}$, apparent rate constant for charge recombination at PS II, leading to formation of ${}^{1}P_{680}^{*}$; k_S , apparent rate
constant for the sum of stable charge separation and
charge recombination, leading to formation of P_{680} or
 ${}^{3}P_{\text{ext}}$ is apparent rate constant fo constant for the sum of stable charge separation and
charge recombination, leading to formation of P₆₈₀ or
³P₆₈₀; *k*_{SV}, apparent rate constant for non-radiative decay
by light-induced Stern Volmer quenchers at PS charge recombination, leading to formation of P_{680} or ${}^{3}P_{680}$; k_{SV} , apparent rate constant for non-radiative decay
by light-induced Stern–Volmer quenchers at PS II; Phe, pheophytin: P electronically excitable ${}^{3}P_{680}$; k_{SV} , apparent rate constant for non-radiative decay
by light-induced Stern–Volmer quenchers at PS II; Phe,
pheophytin; P₆₈₀, electronically excitable component of
PS II^{, 1}P₁₂₂, P₂₂₂ in the (single by light-induced Stern–Volmer quenchers at PS II; Phe,
pheophytin; P_{680} , electronically excitable component of
PS II; ¹P₆₈₀, P₆₈₀ in the (singlet) ground state; ¹P₆₈₀, P₆₈₀
in the singlet excited state; ³ pheophytin; P_{680} , electronically excitable component of
PS II; ${}^{1}P_{680}$, P_{680} in the (singlet) ground state; ${}^{1}P_{680}$, P_{680}
in the singlet excited state; ${}^{3}P_{680}$, P_{680} in the triplet
excited s PS II; ${}^{1}P_{680}$, P_{680} in the (singlet) ground state; ${}^{1}P_{680}^{*}$, P_{680}
in the singlet excited state; ${}^{3}P_{680}^{*}$, P_{680} in the triplet
excited state; [Q_A], the concentration of open PS II
centres: in the singlet excited state; ${}^{3}P_{680}^{*}$, P_{680} in the triplet excited state; [Q_A], the concentration of open PS II centres; [SV], the concentration of light-induced Stern–Volmer quenchers associated with PS II; centres; [SV], the concentration of light-induced Stern–Volmer quenchers associated with PS II; $_{(o)}$ and $_{(c)}$ subscripts applied to rate constants and other terms to signify open or closed PS II centres, respectively Volmer quenchers associated with PS II; $_{(0)}$ and $_{(c)}$ subscripts applied to rate constants and other terms to
signify open or closed PS II centres, respectively (if no
subscript is used, the term applies to open plus closed
centres): ϕ probability of process 'x' occurring centres); ϕ_x , probability of process 'x' occurring; Φ_{x} , yield
of process 'x'. is not closed PS II centres, respectively (if no used, the term applies to open plus closed , probability of process 'x' occurring; Φ_{x} , yield subscript is use
centres); ϕ_x , prof process 'x'.

APPENDIX B

The probability of a photon being re-emitted as **CHLORT ENDIX B**
chlorophyll fluorescence, dissipated through non-radiative
decay or being used to drive charge separation, can be The probability of a photon being re-emitted as
chlorophyll fluorescence, dissipated through non-radiative
decay or being used to drive charge separation, can be
calculated as the rate constant for a particular process chlorophyll fluorescence, dissipated through non-radiative
decay or being used to drive charge separation, can be
calculated as the rate constant for a particular process

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TRANSACTIONS

Triggering of photoinactivation
divided by those for all of the competing processes. For divided by those for all of the competing processes. For
example, the probability of charge separation occurring
within dark-adapted material (when all PS II centres are divided by those for all of the competing processes. For
example, the probability of charge separation occurring
within dark-adapted material (when all PS II centres are
open and ISVI is zero) can be represented by equatio example, the probability of charge separation occurring
within dark-adapted material (when all PS II centres are
open and [SV] is zero) can be represented by equation (B1):

$$
\phi_{C(o)} = \frac{k_{C(o)}}{k_{F} + k_{D} + k_{C(o)}}.
$$
\n(B1)

 $k_{\text{F}} + k_{\text{D}} + k_{\text{C}(0)}$
Because charge separation is a reversible process, the
obability of charge recombination and subsequent Because charge separation is a reversible process, the probability of charge recombination and subsequent transfer of excitation energy back to the pigment hed probability of charge recombination and subsequent transfer of excitation energy back to the pigment bed probability of charge recombination and subsequent
transfer of excitation energy back to the pigment bed
must be taken into account when calculating the yield of
each pathway for de-excitation Equation (R2) expresses transfer of excitation energy back to the pigment bed
must be taken into account when calculating the yield of
each pathway for de-excitation. Equation (B2) expresses
the probability of charge recombination at open PS II must be taken into account when calculating the yield of
each pathway for de-excitation. Equation (B2) expresses
the probability of charge recombination at open PS II
centres once charge senaration has occurred each pathway for de-excitation. Equation (B2) expresses
the probability of charge recombination at open PS II
centres, once charge separation has occurred

$$
\phi_{R(o)} = \frac{k_{R(o)}}{k_{S} + k_{R(o)}}.
$$
\n(B2)

Following charge recombination, the exciton is trans-Following charge recombination, the exciton is transferred to the pigment complex where, once again, it can be re-emitted as chlorophyll fluorescence dissinated Following charge recombination, the exciton is transferred to the pigment complex where, once again, it can
be re-emitted as chlorophyll fluorescence, dissipated
through non-radiative decay or used to drive charge ferred to the pigment complex where, once again, it can
be re-emitted as chlorophyll fluorescence, dissipated
through non-radiative decay or used to drive charge
separation. The reversibility of charge separation must be be re-emitted as chlorophyll fluorescence, dissipated
through non-radiative decay or used to drive charge
separation. The reversibility of charge separation must be
taken into account when expressing the vield of each through non-radiative decay or used to drive charge
separation. The reversibility of charge separation must be
taken into account when expressing the yield of each
pathway for de-excitation. For example, the yield of separation. The reversibility of charge separation must be taken into account when expressing the yield of each pathway for de-excitation. For example, the yield of charge stabilization at open centres in the dark-adapted taken into account when expressing the yield of each
pathway for de-excitation. For example, the yield of
charge stabilization at open centres, in the dark-adapted state, is expressed by equation (B3*^a*):

$$
\Phi_{S(o)} = \frac{k_{C(o)} - k_{C(o)} \times \phi_{R(o)}}{k_F + k_D + k_{C(o)}} \times [1 + \phi_{C(o)} \times \phi_{R(o)} + (\phi_{C(o)} \times \phi_{R(o)})^2 + \dots + (\phi_{C(o)} \times \phi_{R(o)})^n].
$$
\n(B3*a*)

This simpli¢es to equation (B3*^b*):

$$
\Phi_{S(o)} = \frac{k_{C(o)} - k_{C(o)} \times \phi_{R(o)}}{k_{F} + k_{D} + k_{C(o)} - k_{C(o)} \times \phi_{R(o)}}.
$$
\n(B3*b*)

In the light-adapted state, the increase in non-radiative In the light-adapted state, the increase in non-radiative
decay through an increase in [SV] and the presence of
both open and closed PS II centres must be taken into In the light-adapted state, the increase in non-radiative
decay through an increase in [SV] and the presence of
both open and closed PS II centres must be taken into
account For example, the vield of charge stabilization a decay through an increase in [SV] and the presence of
both open and closed PS II centres must be taken into
account. For example, the yield of charge stabilization at
open PS II centres is expressed by equation (B4): both open and closed PS II centres must be taken into account. For example, the yield of charge stabilization at open PS II centres is expressed by equation (B4):

$$
\Phi_{S(o)} = \frac{(k_{C(o)} - k_{C(o)} \times \phi_{R(o)}) \times [Q_A]}{k_F + k_D + k_{SV} \times [SV] + (k_{C(o)} - k_{C(o)} \times \phi_{R(o)}) \times [Q_A]} + (k_{C(c)} - k_{C(c)} \times \phi_{R(c)}) \times (1 - [Q_A]))
$$
\n(B4)

There are four other yields that are of relevance to this study; the yield of charge separation, the yield of fluorescence, the yield of charge recombination, leading to study; the yield of charge separation, the yield of fluorescence, the yield of charge recombination, leading to formation of ${}^{1}P_{680}^{*}$ and the yield of charge stabilization at closed PS II centres. These are given b cence, the yield of charge recombination, leading to formation of ${}^{1}P_{680}^{*}$ and the yield of charge stabilization at closed PS II centres. These are given by equations (B5), (B6) (B7) and (B8) respectively formation of ${}^{1}P_{680}^{*}$ and the yield of closed PS II centres. These are gi (B6), (B7) and (B8), respectively.

$$
\Phi_{\rm C} = \frac{k_{\rm C(o)} \times [\mathbf{Q}_{\rm A}] + k_{\rm C(c)} \times (1 - [\mathbf{Q}_{\rm A}])}{k_{\rm F} + k_{\rm D} + k_{\rm SV} \times [\rm SV] + (k_{\rm C(o)} - k_{\rm C(o)} \times \phi_{\rm R(o)} \times [\mathbf{Q}_{\rm A}]) + (k_{\rm C(c)} - k_{\rm C(c)} \times \phi_{\rm R(c)}) \times (1 - [\mathbf{Q}_{\rm A}]))}
$$
\n(B5)

$$
\Phi_{\rm F} = \frac{k_{\rm F}}{k_{\rm F} + k_{\rm D} + k_{\rm SV} \times [\rm SV] + (k_{\rm C(o)} - k_{\rm C(o)} \times \phi_{\rm R(o)}) \times [\rm Q_A] + (k_{\rm C(c)} - k_{\rm C(c)} \times \phi_{\rm R(c)}) \times (1 - [\rm Q_A]))
$$
\n(B6)

$$
\Phi_{\rm R} = \frac{k_{\rm R(o)} \times [\mathbf{Q}_{\rm A}] + k_{\rm R(c)} \times (1 - [\mathbf{Q}_{\rm A}])}{k_{\rm F} + k_{\rm D} + k_{\rm SV} \times [\rm SV] + (k_{\rm C(o)} - k_{\rm C(o)} \times \phi_{\rm R(o)}) \times [\mathbf{Q}_{\rm A}] + (k_{\rm C(c)} - k_{\rm C(c)} \times \phi_{\rm R(c)}) \times (1 - [\mathbf{Q}_{\rm A}])}
$$
\n(B7)

$$
\Phi_{S(c)} = \frac{(k_{C(c)} - k_{C(c)} \times \phi_{R(c)}) \times (1 - [\mathbf{Q}_{A}])}{k_{F} + k_{D} + k_{SV} \times [\text{SV}] + (k_{C(o)} - k_{C(o)} \times \phi_{R(o)}) \times [\mathbf{Q}_{A}]}\n+ (k_{C(c)} - k_{C(c)} \times \phi_{R(c)}) \times (1 - [\mathbf{Q}_{A}])
$$
\n(B8)

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