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OFFICE OF  
PREVENTION, PESTICIDES AND  
TOXIC SUBSTANCES

**MEMORANDUM**

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**SUBJECT:** Addendum to the Ecological Risk Assessment of Endosulfan

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In 2000, the Environmental Fate and Effects Division (EFED) completed its draft environmental fate and ecological risk assessment chapter (ERA) in support of the reregistration eligibility decision (RED) for endosulfan. The ERA was later revised to reflect public comments and the final version of the science chapter was completed in February 2002 (USEPA, 2002; DP Barcode D238673). At that time, the chapter incorporated nonconservative assumptions in its deterministic aquatic risk assessment and included a probabilistic exposure assessment for aquatic impacts as well.

In addition, because the risk assessment did not take into account the toxicity of the primary transformation product, *i.e.*, endosulfan sulfate, the overall terrestrial and aquatic risk of endosulfan may have been underestimated.

Subsequent to the publication of the final 2002 ecological risk assessment, new information related to endosulfan toxicity, bioaccumulation, monitoring and transport, and ecological incidence have been obtained by OPP. The purpose of this addendum is to conduct a preliminary assessment of this new information in order to: (1) address the extent to which the previous ecological risk assessment for endosulfan might change in relation to this new information, and (2) indicate future

avenues where additional data analysis and risk characterization endosulfan are needed. Areas of focus include new information on endosulfan bioaccumulation, ecological effects, exposure modeling, and monitoring and long-range transport. The findings from each of these focus areas are summarized below. More detailed information in support of these findings is contained in **Appendix 1** and its attachments (under separate cover).

## **EXPOSURE CHARACTERIZATION**

### **Persistence**

Endosulfan is expected to be more persistent than how it was characterized in the 2002 ERA. This conclusion is based on the newly submitted toxicity data on endosulfan sulfate, the major degradate of endosulfan, which shows that endosulfan sulfate is similar in toxicity to the parent compound. Therefore, EFED has used the total residue method to estimate exposure concentrations for endosulfan and endosulfan sulfate. This means that the Agency has based its exposure estimates on the half life of total endosulfan residue, a comparatively longer half life than the parent compound. The total residue aerobic soil half-life is 1336 days compared to the parent only half-life of 57 and 208 days for  $\alpha$  and  $\beta$  endosulfan, respectively.

### **Exposure Modeling**

Revised exposure modeling was conducted based on total endosulfan residues ( $\alpha+\beta$ +sulfate) for two crop scenarios with the highest predicted estimated exposure concentrations (EEC): Florida tomatoes and California strawberries. Predicted surface water EECs increased by 20-40% compared to EECs used in the 2002 ERA, which reflects the addition of endosulfan sulfate in the exposure modeling. Predicted peak, 21-d mean, and 60-d mean surface water EECs are: 23, 9.3, and 6.8 ppb (Florida tomato scenario). Sediment pore water EECs of about 4 ppb were predicted using this same crop scenario and did not vary across these averaging periods. The EECs predicted from the California strawberry scenario are about half those of Florida tomato. However, both California strawberry and Florida tomato EECs may be underestimated because the use of plastic mulch practices, which are expected to increase endosulfan runoff, could not be modeled accurately.

### **Monitoring and Long-Range Transport**

Numerous literature sources have documented the regional or long-range transport of endosulfan through various environmental media such as air, precipitation, water, and sediment. Monitoring studies have detected endosulfan or endosulfan sulfate in environmental media with regular frequency in both agriculturally influenced and remote regions. Recent studies suggest that desorbed residues of endosulfan volatilize and continue to recycle in the global system through a process of migration and redeposition (via wet and dry depositions) as well as air-water exchange in the northern Hemisphere. Findings from selected monitoring studies of endosulfan are summarized below within various environmental media or regions.

#### *Surface Water*

The presence of endosulfan in many ecosystems throughout the U.S. has led to concern regarding continuing point and non-point sources of endosulfan in vulnerable areas that may result in acute and chronic effects on aquatic communities. These data suggest that in the vicinity of row crops where endosulfan is reportedly applied, endosulfan residues have been routinely detected in both the water column and benthic sediments. Additionally, the data indicate that residues exceed OPP acute and chronic risk levels of concern (e.g., south Florida and California) and that total endosulfan residues have moved to areas distant from where it was initially applied. Furthermore, in 1998, Calleguas Creek in California and the Yakima River in Washington State were classified as impaired water bodies under the Section 303d of the Clean Water Act due to presence of endosulfan as well as other chemicals

### *Sediments*

The presence of endosulfan in the sediments of the U.S. is well documented. Based on an analysis of the National Sediment Inventory (NSI) database (EPA-823-C-01-001), between 1980 and 1999, 199 detections were reported for  $\alpha$ -endosulfan (ranging from 0 to 11000  $\mu\text{g}\cdot\text{Kg}^{-1}$ ). For  $\beta$ -endosulfan, there were 667 detections reported (ranging from 0 to 67500  $\mu\text{g}\cdot\text{Kg}^{-1}$ ). For endosulfan sulfate, there were 195 detections reported (ranging from 0.2 to 900  $\mu\text{g}\cdot\text{Kg}^{-1}$ ).

### *Air*

Abundant regional concentration data are available for the Great Lakes Region from the Integrated Atmospheric Deposition Network (IADN), a joint US EPA / Environment Canada-monitoring project (Sun et al., 2003, 3006). These data provide compelling evidence for medium-range airborne transport of endosulfan and endosulfan sulfate, as endosulfan concentrations in vapor phase show a clear increasing trend from the west to east across the region.

### *Remote Regions*

Endosulfan has been routinely detected in remote areas far from sites where it is applied. For example, water samples from four temperate lakes in south-central Canada show the presence of  $\alpha$ - and  $\beta$ -endosulfan (Muir *et al.*, 2004). Mean concentration levels of  $\alpha$ -endosulfan ranged from 28.5 – 1.3  $\text{pg L}^{-1}$ , and those of  $\beta$ -endosulfan ranged from 10.3 – 0.0  $\text{pg L}^{-1}$  in lakes Opeongo, Nipigon, Britt Brook, and Virgin Pond. No agricultural area was within 31 miles (50 Km) of any of these lakes, suggesting that the presence of endosulfan resulted from atmospheric transportation and deposition. Monitoring and modeling results suggest that under the conditions prevailing in south-central Canada, endosulfan can potentially undergo regional-scale atmospheric transport and reach lakes outside endosulfan use areas.

### *Precipitation and Snowpack*

In precipitation of the Great Lakes region,  $\alpha$ - and  $\beta$ -endosulfan concentrations were regularly determined by IADN at various stations during the period of 1987–1997. Concentration levels of  $\alpha$ -endosulfan ranged from 0.13 – 1.95  $\text{ng}\cdot\text{L}^{-1}$  and those of  $\beta$ -endosulfan ranged from 0.19 – 6.09  $\text{ng}\cdot\text{L}^{-1}$  in Lake Superior and Lake Erie. Higher values were reported from Lake Michigan ranging from 0.54

– 8.22 ng·L<sup>-1</sup> for α- and from 1.06 – 12.13 ng·L<sup>-1</sup> for β-endosulfan. Endosulfan and endosulfan sulfate were detected in seasonal snow pack samples at six national parks in the Western United States (Hagman et al., 2006). Concentrations of total endosulfan ranged from 1.5 ng·L<sup>-1</sup> to 0.0040 ng·L<sup>-1</sup> in Sequoia, Mount Rainier, Denali, Noatak-Gates, Glacier and Rocky Mountain National Parks.

### *Mountainous Areas*

The effect of "global distillation" which is believed to account for transport of persistent organic pollutants (POPs) whereby a compound could volatilize from warmer regions, undergo long-range atmospheric transport and subsequently recondense to an accumulation of these substances in the temperate, higher mountainous and Arctic regions.

Aerial transport also caused contamination of snow (Sequoia National Park) and water (Lake Tahoe basin) in the Sierra Nevada Mountains of California/Nevada, a region adjacent to the Central Valley which is among the heaviest pesticide use areas in the U.S. Levels of α-endosulfan found in rain were in a range of < 0.0035 ng/L to 6.5 ng L<sup>-1</sup> while β-endosulfan was determined at concentrations of < 0.012 ng L<sup>-1</sup> up to 1.4 ng L<sup>-1</sup> McConnell et al. (1998).

### *Arctic Region*

Endosulfan has routinely been detected in arctic regions. As excerpted from GFEA (2007):

*“ Endosulfan was reported as a widely distributed pesticide in the atmosphere of Northern polar regions [37]. Unlike for most other organochlorine pesticides average concentrations of endosulfan in the Arctic have not changed significantly during the last five years [38]1. Concentrations of endosulfan (isomers unspecified) from Arctic air monitoring stations increased from early to mid-1993 and remained at that level through the end of 1997 at 0.0042-0.0047 ng/m3.”*

### **Bioaccumulation**

Three lines of evidence were used to evaluate and characterize the bioaccumulation potential of endosulfan and its metabolites: (1) empirical bioconcentration and bioaccumulation experiments, (2) bioaccumulation modeling in aquatic food webs, and (3) bioaccumulation modeling in terrestrial food webs. Based on these lines of evidence, the following conclusions are drawn from this review.

- Empirical data on endosulfan bioconcentration and bioaccumulation are compromised substantially by data quality concerns (i.e., none of the studies appear to meet OPP data quality guidelines) and therefore, BCF and BAF values from these studies should be interpreted with caution.
- Given these caveats, measured BCF values in fish based on the highest quality studies range from about 1000 to 3000 (L/kg wet wt.). This range in BCF values is also supported by

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1 Meakin, S. What's New with POPs Research in the Arctic. *Northern Perspectives* 26 (1), 6-7 (2000)

aquatic bioaccumulation modeling which predict mean and 90<sup>th</sup> percentile BCFs in the range of 1000 to 2000 L/kg wet wt. Considering all studies, BCF values exceeded 5000 for one of seven fish species (yellow tetra), which approximated 5700 (based on the BCF ratio method) and 11,600 (based on the kinetic BCF method). Substantial uncertainties in the BCF values from the yellow tetra study are noted, however, as discussed herein.

- Depuration of endosulfan ( $\alpha$  and  $\beta$  isomers) and its primary degradate (endosulfan sulfate) appears rapid in fish (half life 2-6 days) based on limited data. However, the ability of fish and other aquatic organisms to depurate endosulfan in the environment may be ameliorated in areas of broad-scale endosulfan contamination (e.g., certain agriculturally-influenced ecosystems and even selected remote ecosystems receiving endosulfan loadings through long-range transport).
- Based on one study with daphnids and preliminary aquatic bioaccumulation modeling results, biomagnification of endosulfan appears unlikely in typical aquatic food webs. This finding is consistent with other bioaccumulation modeling studies published in the literature for endosulfan and related compounds.
- Based largely on indirect evidence from terrestrial bioaccumulation modeling, the biomagnification of endosulfan by terrestrial organisms appears to be a concern with predicted biomagnification factors ranging from 2.5 to 28 for herbivorous and carnivorous wildlife at the top of arctic food webs. Additional information on endosulfan in terrestrial food webs should be evaluated to more fully characterize the biomagnification of endosulfan by terrestrial organisms.

## **EFFECTS CHARACTERIZATION**

New toxicity studies from registrant-submitted studies were reviewed and summarized in this addendum. The focus of these studies was exclusively on the toxicity of the primary degrade, endosulfan sulfate. These studies were conducted to address the uncertainty in the relative toxicity of the parent isomers ( $\alpha$  and  $\beta$ ) and the endosulfan sulfate degradate. The new endosulfan sulfate toxicity data consisted of:

- acute toxicity with two species of estuarine/marine fish
- acute and chronic toxicity to one estuarine/marine invertebrate
- sediment toxicity with one freshwater and one estuarine/marine invertebrate
- acute oral and dietary toxicity with two avian species.

Based on comparisons made within the same species, the toxicity of endosulfan sulfate and the parent isomers ( $\alpha$ ,  $\beta$ ) generally appears similar based on acute studies with bluebill sunfish and mallard duck, but somewhat less toxic with bobwhite quail. This information was used to support the assumption that endosulfan sulfate is similar in toxicity to the parent compounds. New information on the toxicity of endosulfan sulfate did not alter the toxicity values used in determining risk quotients for aquatic organisms via water column exposure.

Sediment toxicity of endosulfan sulfate was evident at pore water concentrations as low as 1.5 ppb based on 28-d exposures with an estuarine/marine amphipod.

## **RISK CONCLUSIONS**

Risk quotient (RQ) predicted for aquatic organisms resulting from water column exposure to endosulfan are about 20% to 40% higher compared to the 2002 ERA. This increase reflects the addition of endosulfan sulfate to the exposure modeling and data indicating it is of similar toxicity to the parent isomers ( $\alpha$  and  $\beta$ ). Based on the tomato crop scenario that yielded the largest EECs, acute and chronic RQ values range from about 30 to 60 for freshwater fish, respectively (compared to 23 and 44 from the 2002 ERA) to about 230 and 680 for estuarine/marine fish, respectively (compared to 190 and 490 from the 2002 ERA). Acute and chronic RQ values for invertebrates range from approximately 4 to 130 for freshwater (compared to 3.3 and 93 from the 2002 ERA) and from 50 to 190 for estuarine/marine (compared to 42 to 130 from the 2002 ERA), respectively. Findings from ecological incidents support the findings of endosulfan risk to fish and invertebrates.

Risks to freshwater and estuarine/marine invertebrates resulting from sediment exposure to endosulfan are evident from the integration of exposure and effect characterization. The RQ values for sediment-dwelling invertebrates range from 0.9 to 2.8 depending on species. These RQ values are based on predicted total endosulfan residues compared to endosulfan sulfate toxicity values, which assumes similar toxicity of endosulfan sulfate and total endosulfan residues.

Based on preliminary results from an aquatic food web bioaccumulation model, risks to piscivorous wildlife appear relatively modest with mean predicted acute RQ values exceeding the Agency acute LOC of 0.1 for one of eight species modeled (0.15 for river otter) and 90<sup>th</sup> percentile estimates exceeding the LOC for three of eight species modeled (0.18, 0.39, 0.20 for mink, river otter and belted kingfisher, respectively). Predicted chronic RQ values did not exceed the Agency LOC for any of the eight species modeled.

Risks to nontarget terrestrial wildlife did not change from the 2002 ERA as a result of this addendum, because currently available terrestrial exposure models could not address total residue exposure. Based on the 2002 ERA, RQs for birds and mammals exceed the Agency's acute and chronic risk LOCs and range up to a maximum of 2.7 for birds and 40 for mammals.

In summary, there is a concordance of evidence that endosulfan is undergoing long-range transport and has moved to sites distant from use areas. Additionally, while the parent may readily undergo degradation under some environmental conditions, the sulfate degradate is persistent and represents a source for endosulfan to enter aquatic and terrestrial food chains. While endosulfan is not expected to biomagnify appreciably in aquatic food webs, the compound does bioconcentrate in aquatic organisms to a significant extent. Also, there is direct evidence (measured residues) that endosulfan bioaccumulates in terrestrial systems and indirect evidence (modeling) that endosulfan has a significant potential to biomagnify in certain terrestrial food webs. Monitoring data and incident reports confirm that endosulfan is moving through aquatic and terrestrial food chains and that its use has resulted in adverse effects on the environment adjacent to and distant from its registered use sites.

